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Methods of Measuring Very Long and Very Short Half-lives

Very long half-lives can be measured by the equilibrium and specific-activity methods. Means of determining specific activity are discussed. Pioneer work of Jacobsen and results of others in field of short half-life measurement are outlined

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SHORTLY AFTER the isolation of polonium, Madame Curie observed that the activity of this intensely radioactive substance diminished with time, reaching half its original value after a period of eleven months. In 1900, Sir William Crookes separated from uranium a radioactive substance which he named uranium X. This too was found to lose its activity with time. In 1902, Rutherford and Soddy discovered that an intensely active constituent could be separated from thorium by a single chemical operation. They named it thorium X. A long series of measurements by them showed that the activity of thorium X decays with time according to an exponential law, falling to half its original value in about four days.

A similar exponential decay was observed with other radioactive bodies but the time taken to reach half value was found to vary considerably from one substance to another. These results were generalized into the fundamental law of radioactive transformation by Rutherford and Soddy in 1903. This states that the number of atoms disintegrating per unit time ($-dN/dt$) is proportional to the number of atoms present (N):

$$-dN/dt = \lambda N \qquad (1)$$

where λ is the decay constant. By integration

$$N = N_0 e^{-\lambda t} \qquad (2$$

where N_0 denotes the number of atoms at time $t = 0$. Figure 1 shows a semi-logarithmic plot of this relationship.

Instead of specifying the rate of decay of a particular activity by means of the decay constant, it is usually more convenient to use the half-life T which is the time it takes for half the atoms to disintegrate.

Then $\frac{1}{2}N_0 = N_0 e^{-\lambda T}$

or $$T = \ln 2/\lambda = 0.693/\lambda \qquad (3$$

From the properties of the exponential function it is easy to see that the average or mean life of an atom is $1/\lambda$ and that the probability of an atom existing until time t and then disintegrating between times t and $t + dt$ is $\lambda e^{-\lambda t} dt$.

Since the early days of radioactivity a vast amount of work has been done in measuring half-lives. These values have been found to range from fractions of a microsecond to thousands of millions of years in different elements. Most half-lives are now so well-known that they are the best means for the identification of the radioactivities.

In 1907, Rutherford observed that there seemed to be a relation between the half-life of an α-ray emitter and the

energy of the transformation. Plotting the logarithm of the decay constant against the logarithm of the range of the α particles, Geiger and Nuttall found points distributed almost on straight lines, different for the three radioactive series but parallel to each other. Many years later Sargent showed that a similar sort of relation held for β-ray emitters. Subsequently relations similar to both of these were deduced theoretically on the basis of quantum mechanics. Small deviations between theory and experiment, especially in the case of long or short half-lives, led to a renewed interest in accurate measurements of decay constants and disintegration energies. In most cases the former could be measured much more reliably than the latter, but this was not so for very long and very short periods. Great interest therefore centers round half-life periods in these ranges.

Half-lives of a few seconds up to a few years can usually be measured with sufficient accuracy by conventional methods of timing. The activity, as recorded by some detector, is measured as a function of time and the half-life is calculated on the basis of the equations in the first paragraph. This method fails when the activity shows no sign of decay over a period of many months or when it disappears in a second or so.

VERY LONG HALF-LIVES

One of the longest-lived activities to be investigated by conventional methods is that of europium 154. Fajans and Voigt (*1*) followed the decay of this isotope for 18 months and estimated the half-life as five to eight years. Other workers have followed the 22 year half-life period of RaD by direct measurement. While observations spread over many years would enable even longer periods to be measured, experimenters usually resort to other means

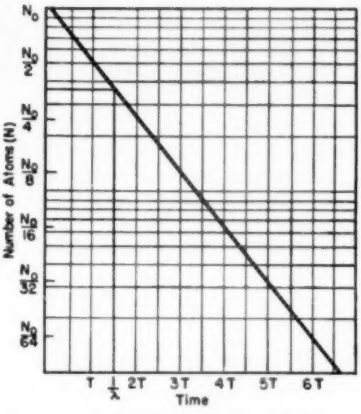


FIG. 1. Graph of fundamental law of radioactive transformation

since they require a result within a reasonable time.

Two methods can be used, although the first mentioned is very limited in its application. It depends on the fact that when radioactive elements are in equilibrium their disintegration rates $(-dN/dt)$ are equal. The other method involves counting the number of disintegrations taking place per unit time in a known number of atoms, and is of universal application.

Equilibrium Method

The use of this method is usually limited by its nature to the natural radioactivities present in minerals of great geological age, where the decay products of some long-lived element have had time to come into equilibrium with the parent. In these circumstances, the disintegration rates of both parent and daughter activities are equal, so that λN in equation 1 is equal for the two elements.

As an example, consider the case of uranium. In minerals of great geological age, this element will be in equilibrium with its later decay product, radium, so that

$$N(\mathrm{U}) \times \lambda(\mathrm{U}) = N(\mathrm{Ra}) \times \lambda(\mathrm{Ra})$$

or if the amounts of the elements present be expressed as weights

$$M(\mathrm{U}) \times \lambda(\mathrm{U}) = {}^{238}\!/_{226} \times M(\mathrm{Ra}) \times \lambda(\mathrm{Ra})$$

Now the ratio of the radium and uranium contents of old minerals has been established chemically to be 3.4×10^{-7} and, since the decay constant of radium is known by other methods, it follows that

$$\lambda(\mathrm{U}) = 4.9 \times 10^{-18}\ \mathrm{sec}^{-1}$$

and the half-life $T(\mathrm{U}) = 4.4 \times 10^{9}$ years.

It would appear that the main use of this method has been in checking values obtained by other means. The period of radium, for instance, can be deduced from a knowledge of the half-life of radon (3.825 days) and the mass of radon in equilibrium with a known mass of radium. The method, however, was very useful in the early days of radioactivity, an early measurement (*2*) of the half-life of RaD, for example, being based on this principle. More recently, Strassmann and Walling (*3*) analyzed mica of known geological age, the strontium content of which is almost pure Sr^{87}. Since this isotope is normally only 7% abundant and since rubidium is also present in the mica, the inference is that all the Sr^{87} is disintegrated Rb^{87}. The decay of the rubidium isotope would be given by

$$N_{\mathrm{Rb}} = N_0 e^{-\lambda t}$$

and the growth of the strontium isotope so formed by

$$N_{\mathrm{Sr}} = N_0(1 - e^{-\lambda t})$$

Thus

$$\frac{N_{\mathrm{Sr}}}{N_{\mathrm{Rb}}} = \frac{1 - e^{-\lambda t}}{e^{-\lambda t}} = e^{\lambda t} - 1$$

The analyses of these experiments enabled the ratio $N_{\mathrm{Sr}}/N_{\mathrm{Rb}}$ to be evaluated, and, since the age t of the mica was known, the decay constant could be calculated. Thus, the value for the half-life of Rb^{87} was found to be 6.3×10^{10} years.

Specific Activity Method

The half-lives of the long-lived radioactivities are more usually determined from observations on the *specific activity* of the element in question, *i.e.*, the number of atoms disintegrating per unit time divided by the number of atoms present. Equation 1 shows this quantity to be equal to the decay constant, from which the half-life can be found.

Measurements of this sort involve counting the particles emitted by a known mass of a radioelement and determining the number of particles emitted in each disintegration. This was first done for radium in 1905 by Rutherford (*4*) who measured the total positive charge carried by the α particles emitted from a known quantity of radium in a known time. It had been shown that each disintegrating atom of radium, freed from its decay products, emitted one α particle. The main uncertainty in the experiment was the magnitude of the charge of the individual α particle. This difficulty was overcome by Rutherford and Geiger (*5*) who devised a method of counting individual α particles. The detector they used was a forerunner of the Geiger-Müller counter but the pulses could only be recorded by an electrometer, setting a very low limit to the counting rate (3–5 per minute). For the determination of the number of α particles expelled from radium, its product RaC was used as a source. The amount of RaC present at any time was measured by comparing its γ-ray activity with that of a radium standard. It was verified that the α particles from the source were emitted equally in all forward directions. Simple geometrical considerations indicated what fraction of the α particles would be detected. In this way, Rutherford and Geiger arrived at a value of 3.4×10^{10} for the number of α particles expelled in one second from

one gram of radium. A knowledge of Avagadro's number enabled the number of atoms in a gram of radium to be calculated; hence the decay constant was determined on the basis of equation 1. Their value for the half-life of radium was 1,760 years.

Perhaps the most accurate experiments of this sort are those of Kovarik and Adams on uranium (*6*) and thorium (*7*). In both cases, the α particles were detected in an ionization chamber and recorded electronically. As in the experiments of Rutherford and Geiger, it was possible to calculate from the geometrical arrangements of the ionization chamber what fraction of all the α particles from the source would be recorded. The experiments on thorium were especially difficult, in that after chemical extraction, thorium is still contaminated with radiothorium and ionium, the latter if uranium was present in the original mineral. To gauge the effect of the radiothorium, it was necessary to know the date of the original chemical separation and, in fact, the experiments were performed at a time when the effect of this contamination was least. The amount of ionium present could be estimated from the radon evolved, since radium and radon are decay products of this isotope. Kovarík and Adams were able to work with samples large enough for precision weighing and they achieved accuracy of a high order in their results. Their value for the half-life of thorium, 1.389×10^{10} years, is the generally accepted one.

In 1939, Nier (*8*) published the results of some accurate measurements made with a mass spectroscope, on the relative abundances of the isotopes of uranium. U^{234} is a decay product of the long-lived U^{238} and in normal uranium they are in equilibrium Now the half-life of U^{238} was accurately known from the results of Kovarik and Adams (*6*), so the results of Nier on the relative concentrations of the two isotopes enabled the half-life of U^{234} to be calculated by the equilibrium method described previously. At the same time, Nier (*9*) made a careful study of the isotopic contents of radiogenic leads (lead formed as the end product of a radioactive series in various minerals), and he came to the conclusion that the actinium series (derived from U^{235}) was only 4.6% as active as the uranium series. This fact can be expressed algebraically in the form

$$\lambda(U^{238}) \times N(U^{238})0.046 = \lambda(U^{235}) \times N(U^{235})$$

Now Nier's previous results (*8*) gave the relative abundances of U^{238} and U^{235}, so that again using the known value of $\lambda(U^{238})$, he was able to calculate $\lambda(U^{235})$ and the half-life (7.13×10^8 years).

In 1946, Chamberlain, Williams and Yuster (*10*) improved on Nier's determination of the half-life of U^{234}. They made a more accurate determination of the abundance ratio using a spectrograph of higher resolving power. At the same time they measured the specific activity of uranium samples enriched in U^{234}. Their results by the two methods are in agreement and their average value is 2.32×10^5 years.

Measurements on the specific activity of long-lived β-ray emitters are generally more difficult than corresponding measurements on α activities because, with these rays, the efficiencies of the detectors are not so easy to determine and the presence of a cosmic-ray background produces an added uncertainty. Bramley and Brewer (*11*) made some careful observations on the specific activity of potassium, determining the efficiency of their counter by using the β rays from a known weight of uranium as a standard. Correction for the absorption of the β rays in the source had to be made and their results are only accurate to about 20%. On the assumption that K^{40} was the

active isotope (*12*), they arrived at a value of 14.2×10^8 years for the half-life.

A few years later Thompson and Rowlands (*13*) showed that potassium has an alternative mode of decay, by *K*-electron capture, transforming into argon. This had been predicted some years before by von Weizäcker (*14*) to account for the abnormally great abundance of A^{40}. In these experiments a potassium salt was placed in a large cloud chamber with a long sensitive time. The great advantage of this method with very weak activities is that radiation from the source can be distinguished, by its appearance, from the cosmic-ray background with most of the latter being effectively eliminated. Photoelectrons of short range were observed in the chamber gas and these were attributed to the characteristic X-rays emitted subsequent to the capture of a *K*-electron by K^{40}. Thompson and Rowlands estimated that this mode of decay was three or four times as probable as β decay. These observations have since been confirmed by some careful work of Bleuler and Gabriel (*15*) using counters. Their value for the branching ratio is 1.9 ± 0.4 and for the half-life $(2.4 \pm 0.5) \times 10^9$ years. Thompson and Rowlands (*13*) pointed out that their own results would indicate a measurable concentration of A^{40} in old rocks containing potassium. Harteck and Suess (*16*) have analyzed the contents of rocks of this sort and on the basis of these results Suess (*17*) concludes that only a few percent of the decaying K^{40} atoms can yield A^{40}. He suggests a half-life of 7×10^8 years as more reasonable than Bleuler and Gabriel's value.

A relation rather similar to that between A^{40} and K^{40} exists for Xe^{129} and I^{129}. The latter isotope has a long half-life of the order of 10^8 years and the unexpectedly large abundance of Xe^{129} can be accounted for assuming its formation as the daughter product of I^{129} (*18*, *19*).

Another long period activity which has been studied recently is that of Re^{187}. Naldrett and Libby (*20*), using a screen-wall counter, were able to detect the presence of soft β rays from this element. There are theoretical reasons (*21*) for assuming that the isotope of mass 187 is the unstable one, so, from a knowledge of its relative abundance and the specific activity of the rhenium, the authors were able to deduce a half-life of 3×10^{12} years.

Another method of determining specific activity involves measurements on the heating effect of radioactivity. Stout and Jones (*22*) have used this phenomenon to evaluate the half-life of plutonium. Their method is similar to that used by M. and Mme. Curie and Sir James Dewar in 1903 in measuring the heating effect of radium (*23*). In the experiments of Stout and Jones, a plutonium sphere was immersed in liquid nitrogen. The rate of evolution of nitrogen gas due to the heat generated by the activity of plutonium was measured. The apparatus was calibrated by the introduction of electrical energy adjusted to give the same rate of evolution of gas. In this way the value of the heat produced could be estimated to 1%. The power generated per atom was calculated and this was combined with the known energy of the Pu^{239} α rays to give the number of α rays emitted per second. The final value of the half-life (2.411×10^4 years) is one of the most accurately known of the long half-lives. The method is not applicable to periods much longer than this because the heating effect would then be too small for accurate measurement.

Photographic plates have recently been used successfully in measuring specific activities. Cuer and Lattes (*24*) dropped some samarium sulphate

solution onto a photographic plate and developed it twenty days later. They were able to estimate the total number of short range α particles emitted in a known time from a known mass of samarium and hence to calculate the half-life $(1.3 \pm 0.1) \times 10^{11}$ years. Yagoda and Kaplan (*25*) used a similar method to check the half-life of uranium and obtained results in agreement with those of Kovarik and Adams (*6*). In a recent paper, Broda (*26*) discussed these methods and asserted that decay constants $\sim 10^5$ times less than that of thorium can be measured in this way. Should this prove the case some new long-lived activities may come to light.

In the case of the artificially produced activities of long life, similar methods are employed but frequently indirect determinations have to be made of the number of radioactive atoms present. Kamen and Ruben (*27*) produced C^{14} by deuteron bombardment of carbon and estimated the total yield as 0.03 μcurie (1 μcurie = 3.7×10^4 disintegrations per second) for 10,000 μampere-hours of deuterons. From these figures, they knew the number of bombarding deuterons but to find the number of disintegrations leading to the formation of atoms of C^{14} they had to rely on rather uncertain estimates of the cross section for $C^{13}(d,p)C^{14}$. Their results gave a lower limit to the value of the half-life at 1,000 years. Reid, Dunning, Weinhouse and Grosse (*28*) were able to improve on these experiments. Having a much stronger activity, they were able to use a mass spectrograph to estimate the abundance of C^{14} in their sample, and knowing the total weight of carbon, they could calculate the number of C^{14} atoms present. Their measurements on the specific activity were subject to considerable corrections, owing to the low energy of the radiations, but they were able to claim an accuracy of 5–10% in their result of 4,700 years. Norris and Inghram (*29*), using a similar method and a higher abundance of C^{14}, arrived at a value of 5,100 years with an accuracy of 4%.

The half-life of Be^{10} has been measured by several experimenters (*30, 31, 32*). In their calculations, they had to rely on rather uncertain estimates of yields and cross sections to obtain the number of active nuclei in the samples they counted. The papers referred to illustrate the roundabout way in which some of these measurements have to be made.

More recently McMillan (*33*) was able to determine with a mass spectrograph the isotopic constitution of an active beryllium sample containing Be^{10} and he estimated the half-life to be $(2.5 \pm 0.5) \times 10^6$ years. Hughes and others (*31*) had arrived at a close value from cross-section estimation.

Finally, in this section on long half-lives, mention must be made of recent work on the period of tritium, H^3. Novick (*34*) collected the He^3 formed by the β decay of tritium and measured its volume. The original hydrogen sample was analyzed for tritium and protium both spectroscopically and by burning with oxygen and weighing the water formed. The sample was allowed to stand for some weeks, and the hydrogen was removed through a palladium valve. The helium was then purified and its volume estimated. Thus by measuring the growth of the daughter product He^3, Novick was able to calculate the half-life of H^3. His value of 12.1 ± 0.5 years has been confirmed by direct measurement on the decay of H^3 by Goldblatt and others (*35*).

SHORT HALF-LIVES

Short half-lives down to about one second can be measured by conventional means, though ingenious methods have frequently been devised to over-

come particular difficulties. For periods much less than this, however, entirely different techniques are required and these will be dealt with first.

The development of these methods can perhaps best be illustrated by the various attempts made to measure the half-life of RaC′. The name of J. C. Jacobsen is associated with much of this work and great credit is due to him for his work in this field of short periods. Interest was centered on RaC′ because agreement with the Geiger-Nuttall relation was found to be best for the uranium-radium series of which RaC′ is a member, and an early extrapolation of this relation indicated a half-life of 10^{-7} second. Jacobsen (*36*) sought a suitably short time interval with which he could compare such a short life and decided to try the recoil of RaC. This atom emits a β ray, recoils as an atom of RaC′ and then, after a time equal, on the average, to the mean life $(1/\lambda)$ of RaC′ emits an α particle. Calculation showed the recoil velocity to be of the order of 10^6 cm per second, so it appeared that some RaC′ recoils could move a measurable distance during their life. Accordingly Jacobsen arranged a source at the end of an evacuated tube and the recoils were allowed to pass through a series of diaphragms. α rays emitted at right angles from the recoil beam were observed at different distances from the source by a scintillation method. The numbers of these were noticed to diminish the farther the detector was from the source. From the results, a half-life of 10^{-6} second was deduced.

Later experiments of Jacobsen (*37*), however, showed that his results were complicated by reflection of the recoil atoms from the walls of the chamber, upsetting the shape of the decay curve. He accordingly set about devising a new method in which two Geiger-Müller counters were used, one to detect the β particles from RaC (heralding the birth of a RaC′ atom) and the other to detect the α particles emitted at the decay of RaC′. If the time intervals between the firing of these counters could be determined, then the mean life of the RaC′ atoms could be evaluated. Subsequent methods of measuring very short half-lives have been based on this principle, differing mainly in the methods of measuring the short time intervals. Jacobsen (*38*, *39*) used a mechanical oscillograph to achieve this. The counter impulses were amplified electronically and the output from each counter fed to a moving-iron mirror-oscillograph. The deflections of the mirrors were arranged to be mutually perpendicular and a beam of light was reflected from each of the mirrors in turn onto a slowly moving photographic film. A source of RaC was placed between the two counters and from the type of trace recorded he was able to deduce that a β ray was frequently followed by an α ray within about 10^{-4} second. Although the type of trace recorded should vary with the time interval between successive particles, the variation in magnitude of the counter pulses prevented an accurate measurement of distribution of relative delay times. However, Jacobsen's value of 2×10^{-4} second for the half-life is quite close to the present accepted value.

The first really accurate determinations of this half-life were made simultaneously and independently by Dunworth (*40*) and Rotblat (*41*, *42*) using similar methods. Rotblat gives a very full account of the experiments in reference *42*. Again two counters were used, one sensitive to β particles and the other sensitive to α particles only. The pulses from the two counters were amplified separately and made of a uniform size independent of the magnitude of the counter pulses. They were then fed into a circuit which

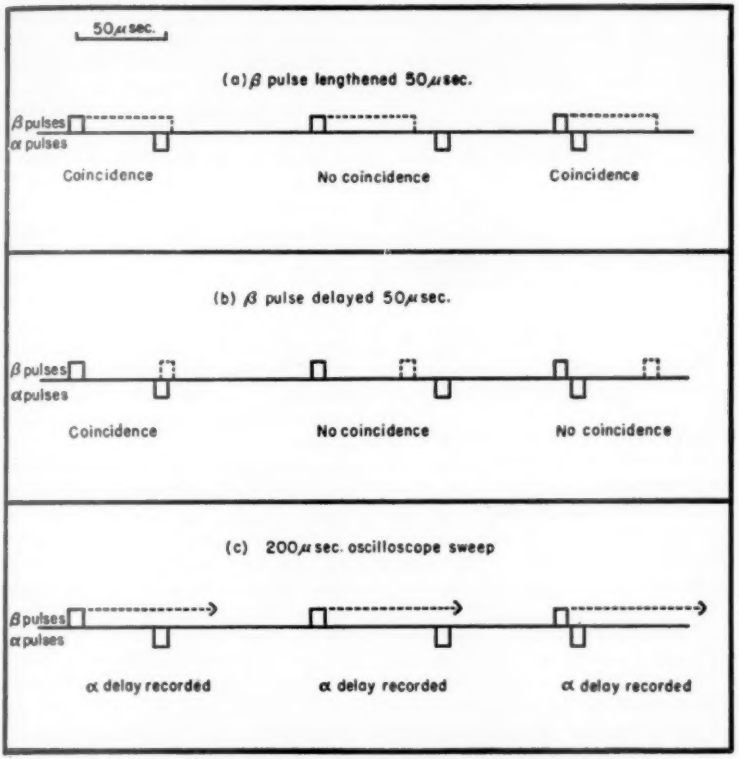
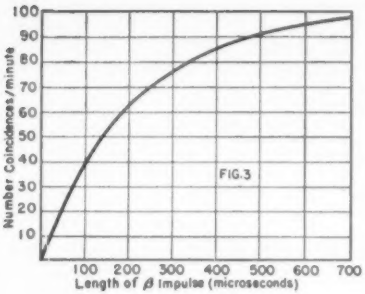


FIG. 2. (a) Pulse-lengthening method (Rotblat, Dunworth, Ward)
(b) Delayed pulse method (Jacobsen and Sigurgeirsson)
(c) Oscilloscope method (Rowlands)

responded only when two pulses arrived simultaneously [Rossi coincidence circuit (*43*)]. Since the pulses have a finite length in time, a circuit of this sort will normally record a coincidence when two particles operate the counters within about 10^{-5} second. However, it is possible to lengthen the first pulse, so that a coincidence is still recorded even when the second pulse arrives after a much larger interval. In Fig. 2*a*, the β pulse is seen to be lengthened to 50 μsec and an α particle arriving in this interval causes a coincidence to be recorded. Thus, Rotblat and Dunworth could, by varying the length of the β pulses, find how many α particles followed β particles within specific time intervals. The type of result obtained is shown in the graph, Fig. 3. It will be seen that this is an inverse of the decay curve for RaC′. If the decay constant is λ, then the probability that an atom will live a time t and then decay in the interval between t and $t + dt$ (counting time since the disintegration of RaC) is $\lambda e^{-\lambda t} dt$. Therefore, the probability for the emission of an α particle from RaC′ during the time τ, or the rate of coincidences



when the β impulse has a length τ, is given by

$$C = C_{max} \int_0^\tau \lambda e^{-\lambda t} dt$$

where C_{max} is the coincidence rate with an infinitely long resolving time.

Integrating,

$$C = C_{max}(1 - e^{-\lambda \tau})$$

showing that the coincidence rate increases exponentially with β-pulse length in a manner defined by the decay constant λ. Rotblat's value for the half-life was 145 $\pm$ 5 microsecond and Dunworth's result was in agreement.

Ward (*44*) introduced a variation on this method by detecting both β and α rays in a single counter and, by an ingenious circuit, measuring the time intervals between pairs of pulses. In the experiments described, he confirmed the previous measurements on RaC′ and also determined the half-lives of AcA (1.83×10^{-3} sec) and ThA (1.58×10^{-1} sec). This method has the advantage that the counter can work at high efficiency, since the source can be inside it, but the analysis of the data is rather tedious. The method will not work for very short intervals between pulses because the counter has to recover from the first pulse before it can detect the one following. This may take 10^{-4} second. Ward admits that the limit of his apparatus was reached with the half-life of RaC′ but there is no doubt that the method could be extremely effective with a detector of short recovery time.

Jacobsen and Sigurgeirrson (*45*) improved on these methods again by making the β and α pulses the same length but delaying the arrival of the β pulses at the coincidence circuit by known times. This is known as the method of delayed coincidences. In Fig. 2*b*, the β pulse is seen to be delayed by 50 μsec and a coincidence is recorded in the first case only. As before, the probability that an atom decays in the interval between t and $t + dt$ is $\lambda e^{-\lambda t} dt$ and if the pulse width is small this is similar to the probability that a coincidence is recorded with a delay t. In fact the graph of coincidences against delay should follow $e^{-\lambda t}$; this was found to be the case. The decay could therefore be observed directly and not by differentiation as in the previous method. In their paper, Jacobsen and Sigurgeirrson establish several advantages of their method over Rotblat's, the most important being a decrease by an order of magnitude in the time taken to obtain results of a similar accuracy. More recently DeBenedetti and McGowan (*46*, *47*) used this method to measure the half-lives of metastable states in Ta^{181} (22 μsec) and Re^{187} (~ 1 μsec).

A further method of measuring short half-lives has been devised by Rowlands (*48*, *49*) and tried on RaC′. This is similar in principle to Jacobsen's oscillograph method (*39*) but a modern high-speed cathode-ray oscillograph is used. Two counters are used; the β pulse triggers the single sweep of the oscillograph and the α pulses are applied to the vertical deflection plates. Provided the time base has been calibrated, the time intervals between pulses can be observed directly by the positions of the α pulses on the screen. If the number of pulses is plotted against the time of appearance after the start of the sweep, the graph is the

decay curve of RaC'. As will be seen from Fig. 2c this method is even more economical in time than the previous one because every α pulse (provided it appears within the sweep time) and its time delay were recorded. Its other advantage is that the apparatus required is available commercially, already calibrated. In its present stage of development, it is not quite as accurate as the other methods, but reliable results can be obtained in a few hour's work. English and others (*50*) seem to have used a similar way of measuring the half-life of At^{217} (21 millisec).

The experiments described above indicate some of the ways in which very short periods may be measured but many ingenious methods have been applied to the determination of periods in the range from say 20 milliseconds to a few seconds. One of the earliest observations on periods of this order was that of Geiger and Marsden (*51*) who noticed that a scintillation from thorium was frequently followed by another within a very short time. They concluded that the thorium emanation must contain two α-ray products, the second of which had an average life of about $\frac{1}{5}$ second. In 1911, Geiger (*52*) found evidence of short-lived activities in the active deposits of thorium and actinium. The periods of these substances, thorium A and actinium A, were determined by Moseley and Fajans (*53*) a short time later. The active matter was deposited from the emanation onto the face of a rapidly rotating metal disk by charging the latter negatively, and the activity of the deposit was examined at different angular distances from the point of deposit by an ionization method. Their results are in close agreement with Ward's measurements made thirty years later (*44*). It is interesting to note that in 1939 Becker and Gaerttner (*54*) used a similar method to measure the half-life of B^{12}. A target of boric acid fused onto an aluminum disk that could be rotated at high speed was bombarded by deuterons. By measuring the activity as a function of angular displacement from the point of bombardment they deduced the half-life of B^{12} to be 0.022 ± 0.002 sec.

With short-lived artificial radioactivities produced by cyclotron bombardment, arrangements have to be made for stopping the bombardment quickly and starting the detection of the induced activities before they have disappeared. Sometimes measurements are complicated by the presence of induced activities in the cyclotron tank and the material under investigation has to be moved rapidly from the site of the bombardment to the detector some distance away. In their experiments on N^{16} (7.3 sec) and He^6 (0.85 sec), Sommers and Sherr (*55*) arranged for the radioactive gases to be released in a target chamber containing two atmospheres pressure of hydrogen to act as a carrier. This chamber could be rapidly opened into an evacuated flow line 120 feet long, and gas at a pressure of one atmosphere could be trapped in a receiver $2\frac{1}{2}$ seconds after bombardment.

Cassels and Latham (*56*) also measured the half-life of He^6 and their result is in good agreement with the value quoted above. In these experiments, a cyclotron was modulated to give short bursts of particles. In between bursts, the counter detecting induced activity was connected in turn for equal time intervals to each of ten recorders. In this way, the fall in counting rate through the ten stages enabled the decay of the activity to be followed. The writers claim that the method can be used from 100 μsec to 1 sec.

Mention should be made of the experiments of Hughes, Dabbs, Cahn and Hall (*57*) on the periods of the delayed neutrons from the fission of

Table of Activities Reviewed

Activity	*Half life*	*Ref.*
Eu^{154}	5–8 y	*1*
RaD	22 y	*2*
Rb^{87}	6.3×10^{10} y	*3*
Th^{232}	1.389×10^{10} y	*7*
U^{234}	2.32×10^{5} y	*8, 10*
U^{235}	7.13×10^{8} y	*8, 9*
U^{238}	4.5×10^{9} y	*6, 8, 25*
K^{40}	2.4×10^{8} y	*11, 12, 13, 15*
	7×10^{8} y	*17*
Re^{187}	3×10^{12} y	*20*
Pu^{239}	2.411×10^{4} y	*22*
Sm^{148}	1.3×10^{11} y	*24*
C^{14}	5,100 y	*27, 28, 29*
H^{3}	12.1 y	*34, 35*
Be^{10}	2.5×10^{6} y	*30, 31, 32, 33*
RaC′	145 μsec	*36, 37, 38, 39, 40, 41, 42, 44, 45, 48, 49*
AcA	1.83×10^{-3} s	*44, 53*
ThA	1.58×10^{-1} s	*44, 53*
Ta^{181}	22 μsec	*46*
Re^{187}	~1 μsec	*47*
At^{217}	21×10^{-3} s	*50*
B^{12}	0.022 s	*54*
N^{16}	7.3 s	*55*
He^{6}	0.85 s	*55, 56*
U^{235} (delayed neutrons	0.05–55.6 s	*57*

U^{235}. The uranium sample was irradiated with neutrons from a pile and then transferred to a well-shielded neutron counter by blowing it down a long tube with compressed air. The delayed neutrons from the sample were then counted and recorded on a moving film. Only about 0.5 second was needed for the transfer and the authors were able to detect six half-lives for the delayed neutrons ranging from 0.05 sec to 55.6 sec.

Finally, it may be useful to draw attention to some experiments of Hipple (*58*) which may have applications to measurements of short radioactive half-lives. He was able to deduce the half-life of metastable ions appearing in mass spectra from the shape of the spectrum peaks in his spectrograph receiver. His undissociated ions were accelerated through a known voltage giving them a known velocity. From the shape of the peaks he was able to deduce at what time during the passage of the apparatus the ions dissociate. He estimated their half-lives to be about 1 μsec.

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Problems in Atomic Energy

DR. O. C. SIMPSON, associate director, chemical division, Argonne National Laboratory, Chicago, at a recent meeting of the American Society of Mechanical Engineers defined eight large general problems in the field of atomic energy as follows:

1. The design and construction of a "more versatile" 60-inch cyclotron in which the type of bombarding particle and its energy can be changed at will.

2. The design of a nuclear reactor for the production of useful power, including the development of a high temperature refractory, such as ceramic materials, for use in place of metals.

3. The engineering of simpler, cheaper, and more versatile apparatus for isotopic separation work.

4. Remote control engineering which "may be the most important single field in the future development of atomic energy." This would include an "atomaton" with apparatus to carry on operations in an air-tight structure heavily shielded against the deadly radiation damage.

5. Problems of shielding which "will play a major role in any use of a nuclear reactor as a mobile power source."

6. Air conditioning and ventilation of radiochemical laboratories, necessary for use with hazardous gases or radioactive particles of matter.

7. New chemical problems which have arisen in dealing with radioactivity.

8. And finally the problem "important not only to atomic researchers, but to every human being, of active waste disposal—sanitation engineering for the atomic age."

Mass Spectrometry and Nuclear Chemistry

The mass spectrometer has become an indispensable instrument in a number of fields of chemistry. Its use in gaseous fission product work is reviewed and some applications to mass spectrometric problems in nuclear chemistry are discussed

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BEFORE 1932, mass spectrometers were used mainly by physicists to identify the isotopes of the elements and to measure their abundances. However, since the discovery of deuterium and the subsequent separation of the light element isotopes, the mass spectrometer has become an indispensable analytical instrument for the chemist.

In recent years, many of these instruments have been built for studying isotopic exchange processes and for use in isotope tracer work (*1–9*). Although radioactive isotopes are now available and are being widely used in isotope tracer problems, the demand for stable isotope tracers, which supplement the radioactive tracers, is steadily increasing. Several commercial concerns are now manufacturing mass spectrometers exclusively for use in this field. Further, the recent development of mass spectrometers, which make it possible to measure accurately the small variations in the isotopic composition of the light elements occurring in nature, has opened up a whole field of investigation.

In the fission of uranium, many isotopes result as products. The process can be represented by the following equation:

$$U^{235} + n \rightarrow [U^{236*}] \rightarrow x_1 A^{m_1} + x_2 B^{m_2} + \nu(n)$$

where $Z_1 + Z_2 = 92$, and $m_1 + m_2 + \nu n = 236$. The primary fission products, A and B, have more neutrons for their charge than the stable nuclei and undergo β-disintegrations in several steps, ending in stable nuclear structures. Some sixty-four such fission chains are now known (*10*).

It was clear from the time of the first fission product studies that the mass spectrometer would eventually be useful in separation problems, mass identification and isotope abundance measurements. However, the early work on fission products involved such small amounts of material that only radiochemical methods were sensitive enough to detect and follow the radioactive isotopes and it was felt that it would be some time before mass spectrometer methods would be practical in this work.

Early in the war program of the Canadian Atomic Energy Project, W. J. Arrol developed techniques for extracting and measuring volumes of less than cubic millimeter amounts of He gas produced in α-emitting substances. These techniques were applied also to the extraction and volume measurement of the noble fission gases (*11, 12*). Later, Thode and Graham found that these small samples of rare gas mixtures (0.001 cc of gas at n.t.p.) were sufficient for mass spectrometer investigations (*13*). Thus, the application of the mass spectrometer to fission product studies became possible for the first time.

The mass spectrometer method provides the only convenient means of identifying and measuring stable isotopes produced in fission. Further, it is the only method of identifying and measuring long-lived isotopes whose activity is too small to be detected by other means. It is the purpose of this paper to review the extensive work which has been done to date with the long-lived and stable isotopes of krypton and xenon arising from the fission of U^{235}, and to point out the importance of the mass spectrometer in these and future nuclear chemical investigations.

Noble Fission Gases

It was well known from early radiochemical investigations of the active fission products that radioactive xenon isotopes are produced in high yield in the slow-neutron fission of uranium-235. Further, several known fission chains were expected to end in stable isotopes of xenon. Interest in these end products led to the development of methods for extracting heavy rare gases from irradiated uranium specimens, purifying them and measuring them in a McLeod gauge, the amounts of xenon expected being of the order of 10^{-3} cc at n.t.p. in uranium samples of about 30 grams.

The gas samples first extracted were found to be much larger than had been expected (*12*). This was explained by the discovery from spectroscopic and mass spectrometric investigations that the samples contained argon. It was later shown that the argon could be extracted from a uranium metal which had not been irradiated. In other words, argon was used as an inert atmosphere in the preparation of uranium metal and was present in all the uranium metal used. Finally, from mass spectrometer studies it was found that the fission gas mixtures contained considerable amounts of fission-product krypton (*13*). The fission gas work, therefore, resolved itself into four sections: the extraction and purification of the rare-gas mixture from neutron-irradiated uranium metal; the separation of these gases, and measurement in the McLeod gauge of the krypton and xenon fractions; and the investigation of the krypton-xenon fractions for radioactivity (long-lived isotopes) At the same time, samples of rare gas mixtures were extracted and purified for detailed mass spectrometric investigation of the krypton and xenon fission products.

Extraction of Fission Gases

Specimens of pure uranium metal are prepared in the form of disks about 1.1 inches in diameter and between 20 and 50 grams in weight. These disks are put together in the form of a rod, sheathed in aluminum and irradiated with neutrons in a pile. To permit most of the fission chains to decay to stable isotopes, the irradiated uranium disks are allowed to stand six months or longer before extraction of the fission gases.

The apparatus used for the extraction is shown in Fig. 1. Saturated potassium cupric chloride solution, freed by air by passage of a stream of pure hydrogen, is transferred to an evacuated flask F_2 where the uranium disks are dissolved to release the fission gases. This vessel is fitted with side arms and ground glass joints large enough to admit a platinum wire basket containing the uranium. The rate of dissolution of the uranium is controlled by raising and lowering the basket to and from the potassium cupric chloride solution. The gas mixture from this dissolution, which consists of a relatively large amount of hydrogen and water-vapour traces of hydrocarbons and small amounts of rare fission gases, is passed through a drying train and finally through a charcoal trap cooled in liquid air, where all gases are ad-

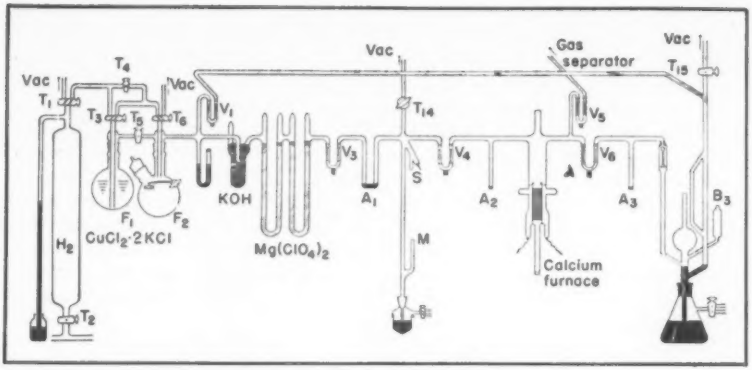


FIG. 1. Fission gas extraction apparatus (*12*)

sorbed except hydrogen which is pumped away through T_{14}.

The gases, now separated from the hydrogen carrier, are manipulated by standard, high-vacuum technique. Mercury ventils replace stopcocks in order to avoid possible loss of xenon and krypton in grease. The mixture of hydrocarbons and rare gases is then transferred into a section of the apparatus containing a calcium furnace similar to that used by Soddy. The gas mixture is subjected to calcium vapour at temperatures between 450° and 500° C for about twenty minutes. Calcium forms solid products with all gases except the rare gases.

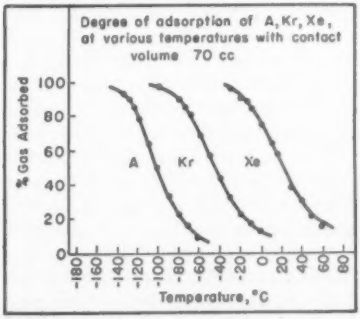


FIG. 2. Adsorption curves for rare gases (*11*)

After this treatment, the rare gases are shown spectroscopicly to be free from hydrogen, hydrocarbons or any other impurities. Samples extracted and purified in this manner are ready for mass spectrometer studies. However, because of the occurrence of krypton and xenon in fission material and of argon in unirradiated uranium metal, it is necessary to separate the gas mixture into the different fractions before quantitative gas measurements with the McLeod gauge can be made.

Separation of Argon, Krypton and Xenon

The method used (*12*) to fractionate cubic millimeter amounts of krypton, xenon and argon depends on variations in the degree of adsorption of these gases on activated charcoal at various temperatures. According to adsorption data obtained for small amounts of each of these gases (Fig. 2), the percentage of argon adsorbed on activated charcoal falls to about 20% at −80° C whereas krypton, which is more strongly held, is nearly 95% adsorbed at this same temperature. Thus for the separation of argon from krypton and xenon, approximately −80° C would be the optimal temperature, and, for the subsequent separation of krypton and

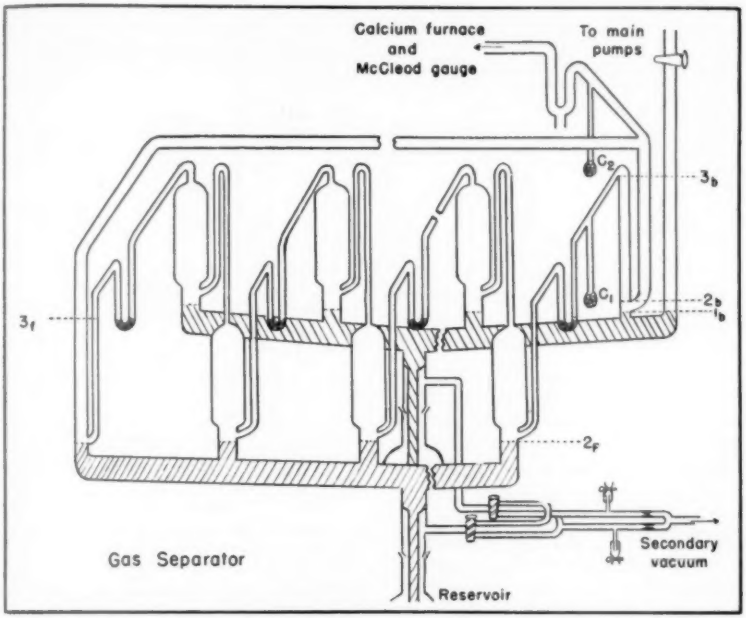


FIG. 3. Apparatus for fractionating argon, krypton and xenon (*12*)

xenon, −20° C would be the optimal temperature.

The separator used to fractionate argon, krypton and xenon (see Fig. 3), is similar in many respects to that used by Gluckauf for the separation of helium and neon. It consists of a series of Topler pumps with U-tubes filled with activated charcoal in every other connecting unit. The pumps are so arranged that the gas phase in unit 1 can be trapped off between pumps and finally transferred to unit 2. In this way, the vapour phase above the activated charcoal will be richer in the gas with the lower degree of adsorption and can be passed on from unit to unit, permitting the usual type of fractionation.

The adsorbed gas and gas remaining in unit 1 can be further fractionated by transferring the vapour phase a second time. This operation is repeated over and over again. Further, at the same time that the vapour phase of unit 1 is trapped off and transferred to unit 2, the vapour of unit 2 is trapped off in the connecting tube and transferred to unit 3, etc. Thus, in nine such operations or transfers, gases appear in the ninth unit. With a charcoal temperature of −78° C and 24 operations of a 9-unit separator, about 95% of the argon can be removed from a ternary, fission-product mixture. By returning the remaining 5% of argon together with krypton and xenon to the beginning of the separator and repeating the process, 99.95% of argon, containing only 1% of the krypton can be removed. In a similar manner, with a charcoal temperature of −20° C, separation of the xenon and krypton fraction can be effected.

TABLE 1

Sample	Weight (gm)	Vol. of gas $\times 10^{-3}$ cc at n.t.p.	Ratio Xe/Kr
Spec. 1	48.2	A 2.80	
		Kr 0.31	5.03
		Xe 1.56	
Spec. 2	135.4	A 3.03	
		Kr 1.03	5.07
		Xe 5.22	

Ratio of Xenon to Krypton

The small xenon and krypton fraction from the separator can be measured with a McLeod gauge. By this method, a cubic millimeter of gas at n.t.p. can be measured reproducibly to an accuracy of ±0.5%. Table 1 shows the amount of argon, krypton and xenon found by W. J. Arrol and his co-workers (*12*) in two specimens of uranium irradiated with slow neutrons.

The Mass Spectrometer

Figure 4 shows an ion source of a 180-degree, direction-focussing mass spectrometer. The direction-focussing principle first used by Dempster (*1*) is illustrated in Fig. 5. Three cases are shown, corresponding to 180, 90 and 60 degree deflection. It may be shown from simple geometry that the resolution, or fractional difference in mass resolvable, is a function only of the radius of curvature of the ion beam and the width of the entrance and exit slits. The angle of deflection is, therefore, immaterial from a resolution point of view (a divergent ion beam of uniform energy V is brought to focus after passing through the magnetic field where the ions are bent according to the relation $e/m = 2V/H^2r^2$, where e is the electronic charge, m is the ion

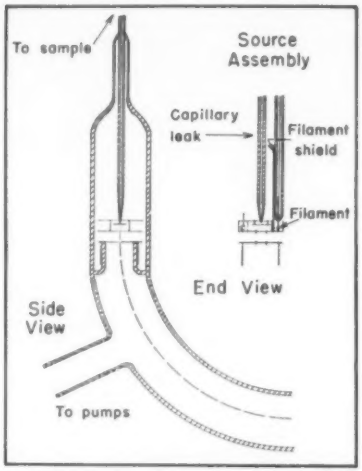


FIG. 4. A 180-deg mass spectrometer ion source

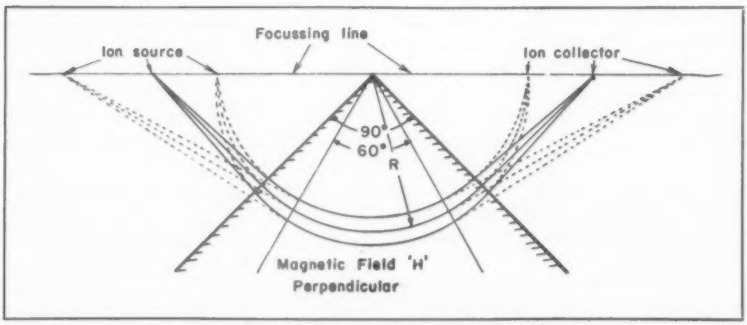


FIG. 5. Direction-focussing method of Dempster

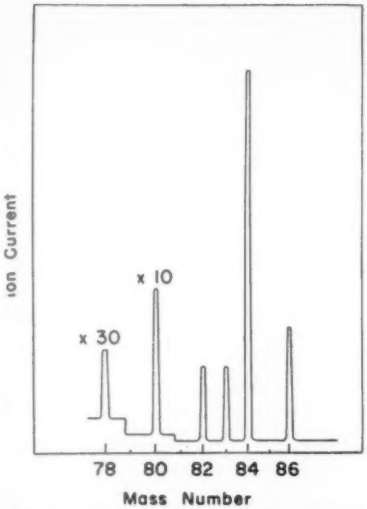


FIG. 6. Recent mass spectrogram of normal krypton from McMaster University (*16*)

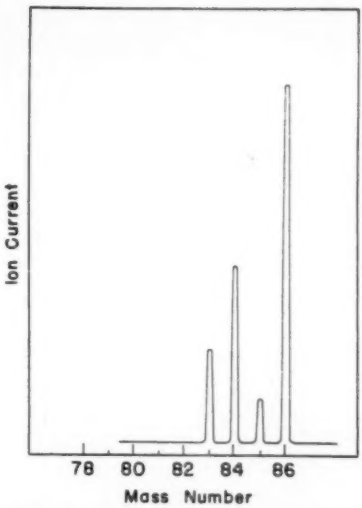


FIG. 7. Recent mass spectrogram of fission-product krypton from McMaster University (*16*)

mass, H is the perpendicular magnetic field and r the radius of curvature).

The mass spectrometer tube may be conveniently considered in terms of an ion source, an analyzer tube and an ion collector. All tubes, regardless of the angle of deflection, can have the same type of ion source and ion collector systems; however, the analyzer tubes must be designed according to the paths indicated in Fig. 5. The instrument is essentially a high-vacuum tube in which gas samples are admitted into the ion source system at low pressures (10^{-4} mm Hg). The molecules of gas are ionized by collision with electrons of controlled energy which travel from a hot filament through a grid to a plate (Fig. 4). The ions so formed will have masses corresponding to the gas atoms and molecules which are present in the sample and to the dissociation products which may be produced in the electron beam. These ions are then drawn from the ionization chamber by a small applied electric field and are further accelerated by a high potential applied between two plates with narrow slits to give a collimated beam. The accelerated beam of ions travels in circular paths as it passes through the magnetic field, the radii of the paths depending on the ratio of charge to mass. By a suitable adjustment of the electrical and magnetic fields, ions of a given charge-to-mass ratio can be focussed through a slit, collected and measured. In actual practice, either the electric or magnetic field is kept constant and the other is varied continuously to scan the different masses. The instrument thus gives direct information on the relative abundance of ionic masses formed when gas is admitted to the tube.

Figures 6 and 8 show typical mass spectrograms obtained using samples of normal krypton and xenon gases, respectively. By the use of automatic scanning and recording devices such as were used to obtain these spectrograms,

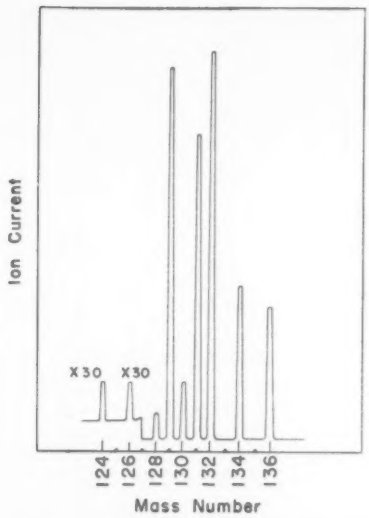


FIG. 8. Recent mass spectrogram of normal xenon from McMaster U. (*16*)

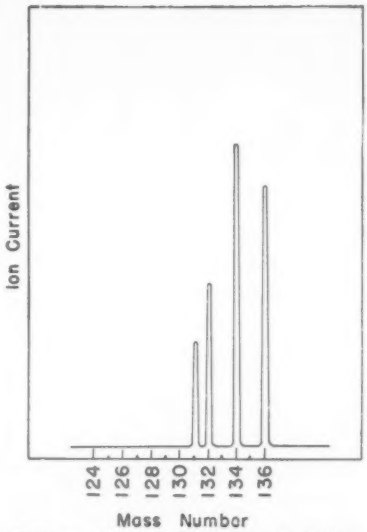


FIG. 9. Mass spectrogram of fission-product xenon (*16*)

the isotope abundances can be measured with a precision of 0.1% (*14*). This is possible even with small fission gas samples where the ion current decreases slowly with time as the sample is used up, since the direction of scanning can be reversed and the double set of peaks can be averaged to give accurate time corrections. See Fig. 10.

The mass spectrometer provides the only convenient means of identifying stable and long-lived isotopes in nature. The application of this method in nuclear chemistry has been particularly useful in the study of fission gases. That there would be stable isotopes of xenon and krypton resulting from the decay of fission-product chains was expected from radiochemical studies of the active members of certain chains. For example, five fission chains had been investigated which could lead to stable xenon. As compiled from Seaborg's table of isotopes (*15*), these are:

					Early branching ratio data
(1)	Sb^{129} 4.2 h	$\rightarrow Te^{129}$ 32 d 72 m	$\rightarrow$ (?)	$\rightarrow$ (?)	0.34
(2)		Te^{131} 30 m	$\rightarrow I^{131}$ 8 d	$\rightarrow Xe^{131}$ stable	1.6
(3)	Sb^{133} 5 m	$\rightarrow Te^{133?}$ 77 h	$\rightarrow I^{133?}$ 2.4 h	$\rightarrow Xe^{133}$ stable	5.2
(4)	Sb^{134} 10 m	$\rightarrow Te^{134?}$ 43 m	$\rightarrow I^{134?}$ 54 m	$\rightarrow Xe^{134?}$ stable	12
(5)			I 1.8 m	$\rightarrow Xe^{-}$	

Similarly four fission chains compiled from the isotope tables could give rise to stable krypton.

(1) $Br^{83} \rightarrow Kr^{83}$
140 m stable

(2) $Br^{84} \rightarrow Kr^{84}$
30 m stable

(3) $Br^{85} \rightarrow Kr^{85} \rightarrow Rb^{85}$
3 m 4 h

(4) $Br^{87}? \rightarrow Kr^{87}?$
50 s

The mass spectrograms obtained for fission-product xenon and krypton, Figs. 7 and 9, can be compared with those obtained for naturally occurring xenon and krypton, Figs. 6 and 8. The marked difference in the number of isotopes present and their abundances is, of course, apparent. Figure 9 shows the existence of four xenon isotopes in fission-product material, masses 131, 132, 134 and 136, and Fig. 7 shows the existence of four isotopes of krypton with masses 83, 84, 85 and 86. Highly purified samples contained less than 1 part in 40,000 of Xe^{129} and Xe^{130} in terms of total xenon and less than 1 part in 8,000 and 5,000 of Kr^{80} and Kr^{82}, respectively, in terms of total krypton. Krypton-85 which does not appear in naturally occurring krypton is radioactive. It is present in all fission-product krypton samples but its relative concentration is less in older samples. The existence of this isotope was not suspected previous to the mass spectrometer investigations because a 4-hour Kr^{85} was already shown to exist and this isotope would have completely decayed in samples six months to a year old. The Kr^{85} found is a long-lived isomer of the 4-hour Kr^{85}. A long-lived isotope of krypton, of unknown mass, was also found by counting experiments (*1*). Its half-life of 9.4 years was measured directly by the mass spectrometer method discussed below.

The two Kr^{85} nuclei mentioned above

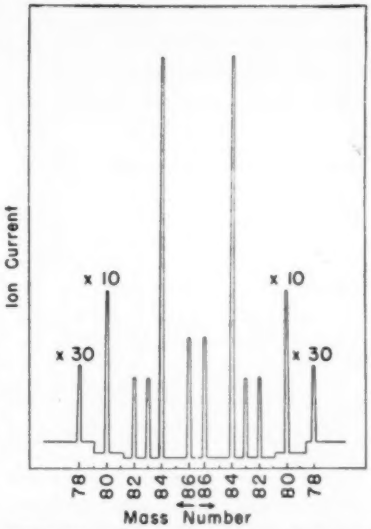


FIG. 10. Mass spectrogram of normal krypton with typical double record obtained by scanning in both directions (*16*)

are daughters of Br^{85} and decay independently to Rb^{85}. Krypton-86 occurs in relatively large quantities although there is no evidence for Br^{86} which would be its parent. This means that either Br^{86} has a very short half-life and has escaped detection, or stable Kr^{86} is formed directly in fission, or both. Since only four isotopes of xenon have been identified with the mass spectrometer, and Xe^{131} is definitely associated with the 8-day iodine, the Xe^{132}, Xe^{134}, and Xe^{136} must be associated with series 3, 4, and 5 above, respectively. The order seems fairly certain since one might expect the iodines to become less stable with increasing mass. Further, fission yield data reported for the above chains should agree with the relative abundances obtained with the mass spectrometer for the corresponding xenon isotopes and, in this way, the mass numbers can be assigned to the various

TABLE 2

Abundance Data

Isotope	*Mass*	*Atom percent*	*Absolute fission yield†*
Krypton	83	14.34 ± .02	0.454
	84	26.72 ± .03	0.847
	85*	7.75 ± .06	0.245
	86	51.19 ± .10	1.623
Xenon	131	13.62 ± .02	2.17
	132	20.30 ± .02	3.23
	134	36.13 ± .03	5.75
	136	29.94 ± .03	4.76

* Krypton-85 radioactive half-life 9.4 y. Percentage obtained by extrapolation to zero time.

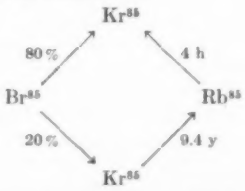
† Calculated, using experimental Xe/Kr ratio = 5.02 and absolute fission yield of Xe^{134} = 5.75 as a standard.

fission-product chains with certainty. Xe^{129} is not present in fission-product samples several years old, at least not in amounts within the limits of detection, *i.e.*, 1 part in 40,000. However, the fission yield for the mass 129 chain is quite considerable. This suggests the existence of a long-lived I^{129} nucleus which, up to the present, has escaped detection. The mass spectrometer could be used to identify I^{129} directly.

Fission Yields by the Mass Spectrometer Method

The relative abundances of stable isotopes of a given element resulting from the decay of fission chains give direct information about the fission yields for these mass chains. In this regard, the fission gases offer a very fruitful field of investigation because of the large number of fission chains (eight) which end in either krypton or xenon. Table 2 gives typical isotope abundance data for fission-product xenon and krypton (*16*). The xenon isotope abundances are reported as percentage of total xenon and those of krypton as percentage of total krypton. The abundances of the xenon and krypton isotopes are then related by means of the ratio of total xenon to total krypton produced in fission. This ratio may be obtained by direct volume measurements of the separated rare gas products or by calibrated mass spectrometer determinations. A Xe/Kr ratio of 5.05, given in Table 1, was used to determine the relative values for column 3, Table 2, and the absolute fission yield of 5.75% for mass 134 was used to convert the values to an absolute scale (*17*).

When these fission yields are compared with the great body of yield data obtained by radiochemical methods and as compiled in the mass-fission yield curve, it is found that six of the values can be fitted to the curve. Actually it would seem logical to draw the curve through these points because of the direct nature of the method used to obtain them and the high precision of the measurements. However, the fission yields obtained by the mass spectrometer method for Kr^{85} and Xe^{136} are 80 and 20% lower, respectively, than the corresponding mass values given by the mass-fission yield curve. In the case of Kr^{85} this discrepancy is easily explained. There are two isomers of Kr^{85} which decay independently according to mechanism:



Mass spectrometer investigations of samples several months old involve the measurement of only the long-lived isomer and the fission yield for Kr^{85} obtained is for the one branch of the chain only. By subtracting this yield from the overall yield for mass 85, read from

the mass-yield curve, one obtains the yield for the other branch of the chain, namely, that through the 4-h Kr^{85}. The yields for the two branches turn out to be in the ratio of 4 to 1 as indicated above (*16*). There is no satisfactory explanation as yet for the low Xe^{136} yield.

An important feature of the mass spectrometer method for determining fission yields is the high accuracy and particularly the high precision obtainable. It is possible with automatic recording equipment to determine isotope abundances with an absolute accuracy of 1% and with a precision of ±0.1%. This compares with errors of ±10% in the values obtained by radiochemical methods. The high internal precision obtainable with the mass spectrometer makes it possible to compare samples extracted from uranium irradiated under different conditions and to measure small significant differences in the distribution of fission-product isotopes. For example, it has already been found that the yields of the noble gas isotopes vary by ½ to 1½%, depending on whether the sample is extracted from a surface or center uranium disk. Actually, one would expect fine structure in the curve for thermal fission yield of U^{235} products as a function of mass number because of delayed neutron emission, coincidental emission, or irregularities in stability of primary fission products. It seems important, therefore, to extend the mass spectrometer investigations to fission isotopes other than Kr and Xe as standard chemical methods are not adequate to show up small variations in the fission yield of a particular chain. However, the mass spectrometer method applied to the rare-gas fission isotopes already provides an elegant procedure for investigating quickly the fission yields of eight isotopes produced by fast and slow-neutron fission of the fissile elements.

Half-life Measurements and Sensitivity of Mass Spectrometer

The mass spectrometer provides a method for the determination of the half-life of a certain radioactive isotope where standard methods may not be adequate because of contamination from other active constituents, or because of difficulties in determining specific activity. For example, the half-life of Kr^{85} was measured mass spectrometrically (*13*). The method involves the determination of the concentration of Kr^{85} relative to a stable isotope and to follow the decrease in this concentration with time. The high precision possible in the determination of relative abundances (0.1% or better) makes reasonably accurate half-life measurements attainable over a considerable range of activities. The half-life of Kr^{85} found by this method is 9.4 ± 0.4 years. It is interesting to point out the micro natures of the mass spectrometer method. The cubic millimeter samples of fission gas at n.t.p. contained less than 10^{-6} cc of Kr^{85}. This constituent could be determined with an accuracy of 1%. The detection limit for Kr^{85} is, therefore, 10^{-8} cc of gas at n.t.p. or about 10^{11} atoms of Kr^{85}. Furthermore, 80% or more of such a sample can be recovered after the mass spectrometer investigation.

Applications of Mass Spectrograph

Some mention should be made of a considerable amount of work which has been done by Dempster, Hayden and Inghram (*18*), *et al.*, to determine mass numbers of radioactive products which appear in fission. For example, a compound is irradiated with neutrons which might give rise to 46-h samarium and 9.2-h europium, both of which are present in fission-product material. A mass spectrograph record is obtained, but, before development, the plate is placed in contact with an unexposed

plate and left for a suitable time. After development, a darkening appears on the unexposed plate opposite the corresponding line which was the source of the activity. If the half-life of the material embedding in the line on the plate corresponds to that of the fission product isotope, then the mass number can be assigned from the position of the line on the plate. The mass numbers of a large number of active products have been assigned in this way.

Nuclear Chemical Problems

It is seen that the mass spectrometer method gives a surprising amount of information about fission-product chains. The various applications to fission product studies are summarized as follows:

1. It is the only direct method for identifying stable isotopes produced in fission.

2. It provides a direct means for assigning mass numbers to the corresponding fission chains.

3. It is the only method of identifying long-lived isotopes whose activity is too small to be detected by other means.

4. An alternative method for the determination of the half-life of certain radioactive isotopes is provided by the mass spectrometer.

5. The mass spectrometer provides a means of determining relative fission yields with high precision. Standard methods do not give adequate precision to measure possible fine structure in fission yield-mass curves.

The extension of the mass spectrometer studies to fission products other than krypton and xenon offers a fruitful field for investigation. However, because of the elegance of techniques for extracting and purifying fission gases and ease of handling rare gases in a mass spectrometer, the fission gases offer the best opportunity for investigating the distribution of fission products obtained under different conditions of uranium irradiation, etc. For example, no study of the yield-mass curve has been reported for fission produced by high-energy neutrons or for change in the fissile nucleus or with the manner of producing fission.

6. Still another application of mass spectrometry to nuclear chemistry and physics is suggested because of the high precision now possible in the determination of isotopic abundances. When a mixture of isotopes is irradiated in a pile with a high neutron flux, the distribution of isotopes will be changed in so far as the isotopes have different neutron capture cross sections. It should be possible, therefore, by means of the mass spectrometer measurements to obtain quantitative data on the neutron capture cross section of the isotopes of certain elements.

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The Liquid-Drop Model for Nuclear Fission

The simple explanation of the mechanism of fission presented by the liquid-drop model is reviewed here with evidence for the quantitatively successful predictions which it provides

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THE LIQUID-DROP MODEL is based on an analogy between certain properties of atomic nuclei and corresponding properties of ordinary matter in the liquid state. The average distance between neighboring particles in the nucleus, the range of the nuclear forces, and the mean de Broglie wavelength per particle are all of the order of 10^{-13} cm. The fact that these three quantities are comparable in magnitude suggests that nuclear matter resembles neither the gaseous nor solid states of ordinary matter but is closest to a quantum liquid.

Weizsäcker (*1*) first showed that the general trend of nuclear binding energies throughout the periodic table could be well represented by the semi-empirical formula

$$E_B = \alpha A \left\{1 - \beta \left(\frac{N-Z}{A}\right)^2\right\} - \gamma A^{2/3} - \frac{3Z^2e^2}{5r_0A^{1/3}} \quad (1)$$

where Z, N, and $A = N + Z$ represent the numbers of protons, neutrons and nucleons respectively.

From the well-known experimental fact that nuclear radii are closely proportional to $A^{1/3}$, it follows that the density of nuclear matter is approximately constant and that the first term in Eq. 1 can be interpreted as a volume energy. This term constitutes the main part of the energy and is to be attributed to the short-range, attractive forces which hold the nucleus together. The part of this term which depends on $(N - Z)^2/A^2$ takes into account the requirements of the Pauli exclusion principle which energetically favors having equal numbers of protons and neutrons.

The second term of Eq. 1 is proportional to the surface area of the nucleus and can be interpreted as a surface tension energy. This term really represents two effects: (1) the desaturation of the short-range cohesive forces at the surface giving rise to a classical surface tension, and (2) an energy correction associated with the diffuseness of the nuclear boundary required by the uncertainty principle.

The third term is the classical electrostatic self-energy for uniform distribution of the protons throughout a sphere of radius, $R = r_0A^{1/3}$. In determining the parameters α, β, γ, and r_0, the observed smooth variation of $N - Z$ with A for stable elements is used to eliminate β and the other three parameters are then obtained by fitting Eq. 1 to measured mass defects. In this way one finds that $\gamma \approx 16$ Mev and $r_0 \approx 1.3 \cdot 10^{-13}$ cm.

The liquid-drop model, which preceded the discovery of the uranium fission, was immediately recognized to provide a simple explanation of the mechanism of fission (*2*). The first term in Eq. 1 is evidently unchanged in a nuclear division in which the protons remain uniformly distributed

throughout the nucleus in the division process, *i.e.*, there is no change in the volume energy associated with the short-range cohesive forces when the nucleus divides. According to the model we use, in which the density is constant or the nuclear matter incompressible, a deformation of the nucleus introduces a change not in its volume energy but only in its shape-dependent surface and electrostatic energies. The normal modes of deformation of the drop are obtained by expanding the radius vector from the center to a point on the surface in a series of surface harmonics.

If we consider only deformation shapes that are symmetrical about the figure axis, the expansion is simply

$$r(\mu) = R[1 + a_0P_0(\mu) + a_1P_1(\mu) + a_2P_2(\mu) + \cdots] \quad (2)$$

where R is the radius of the undistorted spherical drop, μ the cosine of the angle between the radius vector and the figure axis, and $P_n(\mu)$ a Legendre polynomial or zonal harmonic of the first kind. The deformation parameters a_n are the normal coordinates.

The second harmonic P_2 describes an elongation of the drop when a_2 is small and positive; if a_2 is large, the P_2 deformation leads to an equatorial constriction, *i.e.*, a dumbbell shape. Thus P_2 is by itself sufficient to describe a sequence of droplet shapes leading to symmetric fission, and the higher even harmonics P_4, P_6, etc., will be seen merely to fill out some of the sculptural details. A small P_1 deformation displaces the center of the sphere without altering its shape; hence, the shape-dependent electrostatic and surface energies contain no terms in a_1^2. A large P_1 deformation not only shifts the centroid but introduces an asymmetric cardioidal distortion; P_3 tends to produce a pear-shaped deformation. Since these deformations are in general accompanied by a change in the volume of the drop, the term $a_0P_0 = a_0$ must be adjusted to keep the volume constant. Thus a_0 is a known function of the other deformation parameters.

The deformation energy can be simply calculated for small amplitudes. When only quadratic terms are taken into account the deformation energy is found to be

$$\Delta E = 4\pi R^2O \sum_{n=2}^{\infty} \frac{a_n^2(n-1)}{2n+1} \left\{ \frac{n+2}{2} - \frac{10x}{2n+1} \right\} \quad (3)$$

where O is the nuclear surface tension and $2x = (3Z^2e^2/5R)/(4\pi R^2O)$ = electrostatic energy/surface energy of the undeformed drop. The first term in Eq. 3 is the change in surface energy and the second term the change in electrostatic energy. From a purely classical point of view, if the motion of the drop is in accordance with the hydrodynamics of an ideal, incompressible fluid flowing without rotation, the kinetic energy can be calculated to quadratic terms in the time derivatives of the a_n's. Provided that the coefficient of a_n^2 in Eq. 3 is positive, the small amplitude deformations of the P_n variety are simple harmonic and a_n is a periodic function of the time with frequency ω_n given by (*3*)

$$\omega_n^2 = \frac{4\pi O}{3AM} \cdot n(n-1)(n+2) \cdot \left\{ 1 - \frac{20x}{(2n+1)(n+2)} \right\} \quad (4)$$

where M is the mass of a nucleon. If the coefficient of a_n^2 is negative, however, a_n will increase exponentially with the time and the drop will therefore be unstable against small deformations of type P_n. Inspection of Eq. 3 shows that as x is increased, corresponding to increasing charge on the drop, the drop first becomes unstable with respect to second harmonic (P_2) deformations. This instability is reached when $x = 1$, *i.e.*, when the electrostatic energy of the undeformed

drop becomes equal to twice the surface energy. Thus, as the charge on the drop is increased, instability with respect to a particular mode of deformation sets in when the charge is great enough for a small change from the spherical shape to lead to a decrease in electrostatic energy exceeding the increase in surface energy.

Since $4\pi R^2 O = \gamma A^{2/3}$ and $R = r_0 A^{1/3}$, the stability limit $x = 1$ corresponds to $Z^2/A = 10\gamma r_0/3e^2 \approx 48$. Since the heaviest known stable nuclei have values of Z^2/A about ¾ of this critical value, they are consequently stable with respect to small amplitude changes from the spherical shape. If the deformation can be made sufficiently great, however, the decrease in electrostatic energy eventually outweighs the increase in surface energy and, for this critical shape, the drop is unstable against small deformations. A simple calculation shows that the division of a charged drop ($Z^2/A \approx 36$) into two drops in contact is energetically very favorable and that the energy release is a maximum for drops of equal sizes. It would seem evident, then, that instability with respect to P_2 deformations should lead to symmetric fission. This is discussed in the next section.

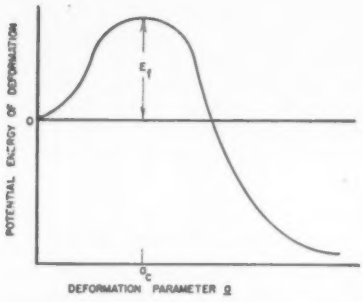
We may summarize the liquid-drop mechanism for nuclear fission as follows: When the charged drop elongates, its surface area and surface energy are increased while its electrostatic energy is decreased because of the spreading apart of the electric charge. For charges less than a certain critical amount (corresponding to $Z^2/A \lesssim 48$ for nuclei), the initial stages of the elongation are accompanied by an increase in surface energy greater than the decrease in electrostatic energy. The nuclear drop is then stable against small deformations but will not be stable against large deformations because the electrostatic energy of an elongated drop decreases more rapidly than the surface energy increases for sufficiently large elongations. It is interesting to note that the stability of a charged liquid drop with respect to small amplitude deformations was investigated by Lord Rayleigh (*4*) in 1882.

The Fission-Activation Energy

The liquid-drop model gives correctly the amount of energy released in the fission process. According to Eq. 1, when the nucleus $_{92}U^{238}$ divides into two equal parts, the energy released is about 190 Mev (assuming the ratio Z/A not to change in the division). A division into unequal parts results in a smaller energy release. More refined calculations give nearly the same results and the agreement with experiment is good. This is less a confirmation of the liquid-drop model than a confirmation of the reliability of the semi-empirical formula (Eq. 1), which can of course be interpreted in terms of the drop model.

A much more severe test of the model is provided in the calculation of the activation energy of fission. If one imagines, for simplicity, that the deformation process is described by a single parameter a and if one plots the potential energy of deformation (surface plus electrostatic) against a, a curve is obtained like that in the figure on the next page.)

When the deformation is small, a can be interpreted as the amplitude a_2 of the second harmonic; at a later stage of the division, a may represent the distance between the centroids of the separating droplets. Since $Z^2/A < 48$ for known nuclei, the spherical shape is one of stable equilibrium and the curve is parabolic near $a = 0$. Because the product nuclei have a smaller potential energy than the initial nucleus, the curve must reach a maximum for a



critical deformation $a = a_c$. The drop is then in unstable equilibrium.

The difference in energy between the critical shape and the initial spherical shape is called the activation energy of fission (E_f). This much energy must be supplied if fission is to take place (apart from quantum-mechanical effects to be discussed later). In the case of slow-neutron-induced fission of U^{235}, the nucleus that undergoes fission is the compound nucleus U^{236} which is formed in an energy state higher than the normal state by the amount of the neutron binding energy. The extra energy may take the form of kinetic energy associated with surface waves (capillary oscillations) of the liquid drop.

If the neutron binding energy is greater than the activation energy E_f, the nucleus will ultimately divide (if a competitive process does not occur). Fission of U^{235} can be produced directly by γ-rays with energies exceeding the value of E_f for this nucleus. The threshold excitation or activation energy will vary from one nucleus to another depending on the value of Z^2/A or x. The smaller the value of these parameters the more stable is the nucleus in question and the larger is the critical deformation and the value of E_f. The main objective of the liquid-drop theory of fission is to predict the dependence of E_f on x or Z^2/A.

The figure opposite naturally oversimplifies the problem. The potential energy is actually a function of all the normal coordinates, *i.e.*, of all the deformation parameters a_n in Eq. 2. Since the primary instability is with respect to P_2 distortions and since the latter describe elongations of the type leading to fission, the parameter a_2 will evidently have the largest value and make the greatest contribution to the deformation energy. Higher harmonics should become successively less important since they correspond to ever more wiggly surfaces (and wiggly integrands of energy integrals whose positive and negative parts tend to cancel). If the critical deformations are not too large, they may then be described with sufficient accuracy by the first few harmonics.

Consider the case in which P_2 and P_4 alone are taken into account. A three-dimensional representation of the potential energy of deformation as a function of a_2 and a_4 is then possible: A potential energy surface is obtained having a bowl at the origin, rising subsequently and falling off steeply for large values of a_2 and a_4. This surface will in general have a col or saddle point corresponding to a path of minimum energy from the region of small to the region of large deformations. The height of the surface at the saddle point is the activation energy.

In analogy with the theory of simple chemical reactions involving a transition state or activated complex, the fission process can be classically symbolized by following the motion of a ball rolling on the potential energy surface. A ball with sufficient kinetic energy to escape may execute many oscillations inside the bowl before it crosses the col. During this time it may lose its energy, corresponding to radiation or emission of a neutron from the compound nucleus. If the ball has just sufficient energy to escape, it will follow the

path of steepest descent beyond the col.

The first detailed consideration of these aspects of the fission process was given by Bohr and Wheeler (*5*). An exploratory calculation was made by them to determine the course of the potential energy surface for small finite deformations in order to estimate the activation energy for uranium fission. They further considered the statistical mechanics of the process and made an approximate estimate of the fission reaction rate. The method used by them to obtain the energy of deformation involves essentially three expansions in series: (*A*) the expansion of the radius vector in the series of zonal harmonics already discussed* (Eq. 2), (*B*) the development of the electrostatic potential in a series of spherical harmonics, and (*C*) the power series expansion of all expressions involving the deformation parameters a_n (which are assumed to be small) giving the deformation energy as a multiple power series in the a's.

There is no difficulty in principle about either (*A*) or (*B*) since only a few of the lowest harmonics are needed in (*A*) and there are a finite number of terms to be taken in (*B*) which depend on the number of harmonics included in (*A*) and the order of approximation used in (*C*). However the uranium isotopes (with $x \approx 0.75$) are not close to the instability limit $x = 1$ and the col of the energy surface must correspond to a fairly large deformation. In the region of interest, then, the deformation parameters are not necessarily small and in particular a_2 would be expected to be large.

Bohr and Wheeler included terms through the fourth order in a_2 and some coupling terms between a_2 and a_4. Higher-order even harmonics and the odd harmonics were not taken into account. An extension (*6*) of these calculations to higher-order terms showed that the multiple power series was very slowly convergent and that the few terms considered by Bohr and Wheeler, while valid in the immediate neighborhood of $x = 1$, were insufficient to give accurate results at $x = 0.95$ and became totally unreliable for $x = 0.9$. Bohr and Wheeler made a further approximation which tended to cancel the error incurred through the inadequacy of their power series; therefore, their activation-energy curve, while reasonable as a semi-empirical approach, is not based on an accurate solution of the liquid-drop problem.

Present-Reines-Knipp Investigations

The most extensive investigations of this problem since the pioneering work of Bohr and Wheeler are those of Present, Reines, and Knipp (*7*) and Frankel and Metropolis. (*8*). The method developed by the former authors (PRK) avoided the expansion in terms of the large parameter a_2 but maintained the expansions in the other small parameters (a_4 and a_6) and the development of the potential in spherical harmonics. Separate calculations were made for $a_2 = 0.3$ and 0.4 and the results of the power series method were used for smaller values of a_2. This made possible an accurate determination of the activation energy and the saddle point deformation parameters for nuclei in the range $0.8 < x \leq 1$.

Extrapolating these results to the compound nucleus U^{239} with $x = 0.74$, the theoretical value for the activation energy E_f was found to be 7.4 ± 0.7 Mev where the error indicates the uncertainty introduced by the extrapolation. This error is smaller, however, than the probable error in x and E_f arising from uncertainty in the coefficients of the binding energy formula

* Bohr and Wheeler made their calculation in spheroidal coordinates and used a_2 to denote the amplitude of a purely spheroidal deformation.

(Eq. 1).* The experimental value of E_f for the U^{239} fission obtained from the estimated neutron binding energy and the threshhold for fast neutron fission of U^{239} is approximately 6 Mev.

Frankel-Metropolis Calculations

Subsequent calculations by Frankel and Metropolis (FM) confirmed these results. In their work no expansions were made beyond the original representation of the radius vector as a series of zonal harmonics. The integrations were carried out with the aid of the large electronic calculator known as the Eniac. In this way Frankel and Metropolis were able to treat large deformations beyond the range of the expansions used in the earlier work. The two sets of calculations agree precisely at $x = 0.9$; the FM result for E_f at $x = 0.8$ is about 6% higher than the PRK value. At $x = 0.74$, the Eniac gives $E_f/4\pi R^2O = 0.0136$. It may be concluded that the agreement between the theoretical value predicted on the basis of the liquid-drop model and the value inferred from experiment is well within the uncertainties resulting from our inexact knowledge of the nuclear constants.

The calculations described above with even harmonics only correspond to a symmetric fission. Experiments indicate, however, that fission is in all cases preponderantly asymmetric. Deformation shapes leading to asymmetric fission are described by the odd zonal harmonics. The multiple power series of the PRK calculation included the lowest-order terms in a_1 and a_3 and their coupling terms with a_2. These terms were found to make a positive contribution to the deformation energy and, extrapolating the results to uranium, it seemed likely that the saddle point of the energy hypersurface (ΔE versus a_1, a_2, a_3, a_4, etc.) would correspond to a symmetric deformation shape. Similar conclusions were reached by Frankel and Metropolis from a more thorough investigation of the large amplitude distortions.

Since even the latter calculations cannot be extended to deformations so large that the radius vector intersects the surface twice, the possibility has not been excluded that for very large deformations the contribution of the odd harmonics may change sign and the path of steepest descent beyond the saddle eventually deviate from the axis of symmetric deformation shapes. This supposition seems very unlikely, however, because the deformation shape for large distortions is well-approximated by two drops in contact. Two charged drops of equal sizes in contact have, however, a lower energy than two of unequal sizes resulting from the same original drop. Thus it seems unlikely that the asymmetry of fission can be accounted for on the basis of the liquid-drop model.

It may then seem questionable to compare the calculated value of the activation energy for a symmetric distortion with the experimental value which corresponds to a markedly asymmetric division. It is possible, however, that the critical shape of the real nucleus is symmetrical and that the asymmetry develops in the later stages of the division process beyond the col of the energy surface. Even if this is not the case, the critical shape, which occurs in the early stages of the division for uranium, must be very close to symmetry and the activation energy cannot therefore be seriously in error.

In regard to the approximation made in the above purely classical treatment by neglecting quantum effects, the amplitude of the zero-point oscillations is, in fact, small compared to the nuclear

* The values of the nuclear constants used were: $\gamma = 16$ Mev, $10\gamma r_0/3e^3 = 48$ and $4\pi R^2O = \gamma A^{2/3} \cong 600$ Mev for uranium.

radius, and the zero point energy of the fundamental mode $\hbar\omega_2/2 \approx 0.4$ Mev is a small fraction of the activation energy. This ensures the approximate validity of a classical description. No correction is made for the zero-point energy in the above calculations, since the drop in the activated state has also a zero-point energy of uncertain amount, and it would seem more consistent to neglect zero-point effects altogether.

A second quantum effect which must be taken into account is the finite probability of fission occurring when there is insufficient energy to surmount the potential barrier. In analogy to the radioactive alpha decay in which a nucleus divides spontaneously into an alpha particle and residual nucleus via a path through a region in which its kinetic energy would be classically negative, it is possible for a nucleus with $x < 1$, which is classically stable against small deformations, to undergo spontaneous fission by quantum-mechanical barrier penetration or tunnel effect.

It is possible to estimate the rate of spontaneous fission on the basis of liquid-drop calculations (*7*, *8*). The penetration probability is greatest for a path through the many-dimensional barrier corresponding to an irrotational motion of the nuclear particles along classical streamlines (motion of minimum kinetic energy). The assumption of irrotational motion, together with a knowledge of the potential energy hypersurface obtained from the preceding calculations, enables one to calculate the Gamow penetration integral for the spontaneous fission. The results depend very sensitively, of course, on the width and height of the barrier and, therefore, on the value of x for the nucleus in question.

The computations of Frankel and Metropolis give a mean lifetime of about 10^{26} years at $x = 0.74$ and 10^{17} years at $x = 0.75$. In view of the uncertainties in the nuclear constants which affect the value of x, these results are seen to be compatible with measurements (*9*) of spontaneous fission in natural uranium which indicate a mean lifetime of about 10^{17} years.

Summary

The main results may be summarized as follows: (1) The liquid drop model provides a clear and simple picture of the mechanism of nuclear fission. (2) The model is quantitatively successful in predicting the activation energy for fission and the lifetime against spontaneous fission. (3) The model is unable to account for the observed asymmetry of fission.

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Radioisotopes in Nutritional Trace Element Studies—I

First of a series of three articles presenting techniques and results of radiotracer studies in plant and animal metabolism

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THE TERM "TRACE" was originally used, in the chemical sense, to designate an amount of a constituent so minute that it could not be quantitatively measured. In recent years, coincident with the development of more sensitive chemical analytical methods, this expression has come to signify a minute amount of a constituent which it may be necessary to determine and which can be determined. In biochemistry, the terms "trace elements," "micronutrients" and "minor elements" are used interchangeably to specify certain chemical elements which are essential in the metabolism of plants and animals and are required in only extremely small quantities. This usage should not be confused with such expressions as "tracer studies," which signify the use of isotopically labeled substances to follow the fate of a given element or compound. The meaning will usually be clear from the context.

The availability of powerful analytical methods, based primarily on photoelectric spectrophotometry, filter photometry, and spectrography has led to a beginning in understanding the role and function of trace elements in the life processes. Nevertheless, chemical methods have not been able to supply the basic information as they have for the major elements and compounds of biochemical importance; this is due primarily to the inherent limitations which assume increasing significance the smaller the amount of the element physiologically involved. These limitations arise chiefly from:

(*A*) the sensitivity of the chemical methods—usually about 10^{-4} per cent, which may be low compared with the amount of element necessary to measure;

(*B*) the difficulties in determining a minute quantity of an element in the presence of an overwhelming quantity of other substances;

(*C*) the impracticality of raising the level of the element in a tissue by a determinable amount, without resorting to massive or non-physiological doses;

(*D*) the time and labor necessary to obtain reliable analytical values.

It has become apparent that radioisotope procedures offer relief from many of these limitations and are, therefore, uniquely suited to investigating interrelationships and overlapping factors in trace element metabolism. They are particularly effective in combination with established chemical and biochemical procedures.

This review* is limited to considering nutritional trace element studies in animals, although many of the methods discussed are equally applicable to plant investigations. Discussions of technique are presented from the point of view of the investigator in the relatively small laboratory who may wish to use radioisotope procedures in conjunction with other methods and

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who is, therefore, interested in employing convenient and commercially available equipment whenever possible.

The Trace Element Problem

There are several excellent reviews (*1*, *2*, *3*, *4*, *5*, *6*), which may be consulted for full details of the status of trace-element research. The general statement of the problem which follows is intended merely to provide an understanding for the increasing use and need of radioisotopes in this field of investigation. In later sections of the paper, the individual trace elements will be considered in more detail.

The substances necessary for normal animal health can be classified according to their main function. The major dietary essentials, such as fats, carbohydrates and proteins are required in large amounts and serve to supply energy for body activities and structural materials for growth and upkeep of tissues. Certain minerals, particularly calcium and phosphorus are also required in large amounts primarily for structural purposes. On the other hand, such compounds as the vitamins are required only in minute amounts, since their function is concerned mainly with the regulation of the body processes. Within recent years there has been general recognition that certain minerals in extremely small amounts are essential for animal health, having a regulatory function similar to that of the vitamins. The minerals considered in this review are copper, cobalt, molybdenum, manganese and zinc.

There are two very good reasons why observations on farm animals have been of importance in bringing to light the fact that trace-element inbalance may lead to disease. In the first place, livestock are often restricted to forage from a relatively small area, and this increases the chance of development of disease symptoms if the area should happen to be deficient. With man or animals getting food produced in various sections of the country, any deficiency in that from a given area will be at least partially compensated in the total diet. The other point is that ruminants, as a result of their peculiar digestive processes, have been observed to require a higher level of certain trace elements than do other species. In general, it is considered that mineral deficiencies are more important than vitamin deficiencies as far as the grazing animal is concerned. This accounts for the emphasis placed on trace-element studies with ruminants, not only from the point of view of improved animal health, but also as a logical starting point for investigating the role of these elements in the nutrition of all species.

There is abundant evidence (*7*) that malnutrition in general may be caused by conditions other than a primary dietary deficiency. Factors that interfere with the absorption and utilization of nutrients, or those that increase their destruction and excretion, must be taken into account. Body requirements may be increased by detoxification processes, physical exertion, fever, drugs, toxins, abnormal environmental conditions, pregnancy, lactation and any related conditions that result in an increased metabolic requirement on the part of the tissue cell.

In regard to the role and behavior of trace elements in animals, it has become clear that interactions between elements and compounds are of considerable significance. The most obvious type of interaction is the altering of the solubility of a substance in the gastrointestinal tract. As an example, absorption of phosphorus is diminished by factors that favor the formation of insoluble salts of phosphoric acid; the administration of an excess of such cations as beryllium, iron, and aluminum will inhibit the absorption of the phosphorus and lead to the development of

TABLE 1

Availability of Radioisotopes for Trace Element Metabolism Studies

Item No.	Isotope	Half-life	Radiation Energy (Mev) Beta	Gamma	Production	Projectile and Target
1	P^{32}	14.3 d	1.69		Pile	*n*, S
2	Ca^{45}	180 d	0.25		Pile	*n*, $CaCO_3$
2a	Ca^{45}	180 d	0.25		Pile	*n*, CaO
3	Mn^{54}	310 d		0.85	Cyclotron	*d*, Fe
4	Fe^{55}	~4 y	*K*	0.07	Cyclotron	*d*, Mn
5	Fe^{59}	44 d	0.26 0.46	1.1 1.3	Cyclotron	*n*, Co (Fast neutron)
6*	Fe^{59}	44 d	0.26 0.46	1.1 1.3	Pile	*n*, Fe
6a*	Fe^{59}	44 d	0.26 0.46	1.1 1.3	Pile	*n*, Fe (Long irradiation)
	^{56}	85 d	1.4 β^+	0.85, 1.27, 2.6, 3.3	Cyclotron	*d*, Fe
	^{57}	270 d	0.26 β^+	0.12, 0.13, *K*		
7	Co^{58}	65 d	0.4 β^+	0.81, *K*		
8	Co^{60}	5.3 y	0.3	1.1, 1.3	Pile	*n*, Co_2O_3
8a	Co^{60}	5.3 y	0.3	1.1, 1.3	Pile	*n*, Co_2O_3 (Long irradiation)
9	Cu^{64}	12.8 h	0.58 β^- 0.66 β^+ *K*	1.2 (weak)	Cyclotron	*p*, Ni
10	Cu^{64}	12.8 h	0.58 β^- 0.66 β^+ *K*	1.2 (weak)	Pile	*n*, Cu
11	Zn^{65}	250 d	0.4 β^+ (1%) *K*, e^- (99%)	1.14	Cyclotron	*d*, Cu
12	Zn^{65}	250 d	0.4 β^+ (1%) *K*, e^- (99%)	1.14	Pile	*n*, Zn
12a	Zn^{65}	250 d	0.4 β^+ (1%) *K*, e^- (99%)	1.14	Pile	*n*, Zn (Long irradiation)
13	Mo^{99}	67 h	1.3	0.77, 0.815 0.840	Pile	*n*, MoO_3
14	I^{131}	8 d	0.6	0.367, 0.080	Pile	*n*, Te

* Pile bombardment of iron produces both Fe^{59} and Fe^{55}.
† A preparation obtained from MIT; specific activity expressed as milligrams of radium gamma-ray equivalent per gram.
‡ A preparation reported in (*17*).
§ Available as metallic Co by special arrangement.
‖ No carrier added.

Form Available	*Specific Activity Initially Available (mc/gm)*	Cost ($ per mc)
Na_2HPO_4	N.C.A.‖	1.10
$CaCO_3$	0.1	41.20
$CaCl_2$	28	41.20
Fe	0.06	33.00
Fe	0.25	33.00
	10.7×10^3 †	
Co_2O_3	30	1.65
Co_2O_3‖	250	1.65
	4000‡	
Cu	300	0.12
Zn	1.9	2.20
Zn	25	2.20
MoO_3	10	0.30
	N.C.A.‖	1.70

characteristic rickets in the animal (*8, 9*). Reduction of the toxic effect of soluble copper by drying it onto the food represents another instance of this behavior (*10*). Less obvious mechanisms are involved in such relationships and antagonisms as copper vs. molybdenum (*4*), cobalt vs. histidine (*11, 12, 13*), cobalt vs. sulfur-containing amino acids (*14*), and manganese vs. organic antiperotic factors (*15*).

In the investigation of any single element, the need to consider the effects of numerous other elements and compounds leads to great complexity. This accentuates the necessity for simple techniques by means of which large numbers of experiments can be performed to separate out the effect of all important variables. Such observations as the possible relationships of copper, molybdenum and manganese to bone development, and the role of copper in iron utilization call for use in these studies of the radioisotopes of phosphorus, calcium and iron in addition to those of the trace elements themselves. As the locational behavior is clarified and the experiments logically develop to functional aspects, the isotopic labeling of organic intermediates will become of increasing interest in this field.

Availability of Radioisotopes

Table 1 presents a list of the radioactive isotopes found most useful in this type of investigation. All known radioisotopes of the given elements are not listed, nor all of the reactions which could be used for their production; only those deemed most suitable for nutritional trace-element studies are described. Information on the pile-produced isotopes was compiled from the literature furnished by the Isotopes Division of the AEC, whereas the data on the cyclotron-produced isotopes were taken mainly from Irvine (*16*) except in the cases of items 7 and 9,

which represent specific preparations.

The specific activity of the pile-produced isotopes has been expressed in millicuries per gram where the millicurie has been defined as 3.7×10^7 disintegrations per second or, in the case of an indeterminate mixture of isotopes, as 3.7×10^7 beta emissions per second in the sample, in accordance with AEC usage. In the case of item 7, a mixture of cyclotron-produced cobalt isotopes, the specific activity has been expressed in milligrams of radium gamma-ray equivalent per gram as suggested by Evans (*18*). The specific activity values depend on many factors and may vary from one preparation to the next so that the values given are to be regarded only as estimates. Items 7 and 9, cobalt and copper, represent individual preparations and the specific activities are listed only to show what can be done and for comparisons of order of magnitude with the specific activities available from nontransmutation reactions. Cost figures for the pile-produced items have been established by the AEC and can be used to ascertain the scale of experimentation feasible. At the present time, the Committee on Nuclear Science, National Research Council, is making an effort to initiate a coordinated program for the production and distribution of cyclotron-produced isotopes through the facilities of the AEC or other appropriate organization. However, there is as yet no basis for cost estimation. It might be expected in the case of Cu^{64}, item 9, that the total activities available would be relatively small, so that only in small scale experimentation would the advantage of the high specific activity become of actual benefit.

Considerations of Dosage

The dosage of a labeled element administered to an animal must be considered both from the point of view of the radioactivity present and the total amount of the element involved. The definitions below are concerned with total amount of element administered, it being assumed that there is sufficient radioactivity to make measurements throughout the experiment:

Tracer dose: *The amount of the administered element is small compared with the normal intake.* **Physiological dose:** *The amount of the administered element is of the same order of magnitude as the normal intake.* **Massive dose:** *The amount of the administered element far exceeds the normal intake. When the level of radioactivity in the administered dosage is sufficient to produce a biological response, the term* **therapeutic dose** *has been used.*

It is considered that the tracer dose and physiological dose will follow the normal path, since the amount of the element introduced into the system will not be large enough to disturb equilibrium or steady-state conditions. On the other hand, a massive dose may well flood the tissues and indicate an abnormal distribution. The classic example of flooding is the behavior of radioiodine administered to rabbits, as reported by Hertz, *et al.* (*19*). The percentage collection of the iodine dosage by the thyroid was directly affected by the amount injected; the percentage collection increased as the dosage was decreased. In terms of a concentration coefficient, which was a measure of affinity of the thyroid for iodine, the value at 0.1 mg dosage was about 80, while for an injection of 100 mg it was found to be 2 or less. In a study of toxicity, however, it would be desirable to use a near-toxic level. This usually presents no problem since the dosage can be increased merely by using an additional inert element. Difficulties usually arise when it is necessary to keep the dosage at a minimum. An example of a thera-

peutic dose would be the use of sufficient radioiodine to bring about destruction of thyroid tissue.

The ideal tracer experiment consists in the use of a tracer dose, or if that is not possible, a physiological dose with a level of activity such that a convenient size sample gives a count several times that of the counter background at time of measurement; no radiation effects would be expected at this level of activity except perhaps with short-lived isotopes where a high initial activity may be required, or in a case where the accumulation in the particular tissue of interest is very low. In practice it is usually necessary to make a compromise depending largely upon the characteristics of the isotope preparation.

From the data in Tables 1 and 2, it is possible to estimate the dosage required for a given experiment, and also the cost in the case of AEC isotopes, provided information is available on the approximate accumulation of the isotope in the tissues to be studied.

Measurement of Radioactivity

The measurement of low-beta-energy emitters, as for instance Fe^{59}, Ca^{45}, and Co^{60} of the elements listed in Table 1, is a considerably more tedious and difficult operation than the assay of the higher-energy emitters; this is also true for Fe^{55}, which has a very soft X-radiation. In general, with the soft-beta emitters it is necessary to remove the desired element from the extraneous accompanying material either by electroplating or precipitation as a chemical compound, after which it can be measured with a thin-window Geiger counter or electroscope, with great attention being paid to the geometry and corrections for self-absorption. A proportional counter has recently appeared on the market which is claimed to have certain advantages over the conventional mica window counters for this purpose.

It is fortunate that most of the radioisotopes of interest in nutritional trace element studies have sufficiently high-energy radiation so that their assay becomes relatively simple. The measurement of soft emitters will not be considered further since this subject is beyond the scope of this review and has been extensively treated in the literature [general considerations (*20*, *21*, *22*), carbon-14 (*23*, *24*, *25*), sulfur (*26*), calcium (*27*, *28*, *29*), cobalt (*30*, *31*), iron (*32*, *33*)]. Although the measurement of the higher-energy radiation has been taken largely for granted, the procedures described below and found convenient in use by the Florida Agricultural Experiment Station may be of interest to other workers.

It has been satisfactory, from the standpoint of sensitivity and reproducibility, to make all assays of the high-energy emitters in solution. The geometry is easily reproducible and the preparation of the sample for assay is a very simple and rapid operation. This is in contrast to the counting of solid samples in which case it is necessary to prepare samples with a flat and uniform surface, and to calibrate the self-absorption effects. In solution measurement, when samples contain too much activity it is a simple matter to dilute or take an aliquot for assay.

Three types of commercially available Geiger-Müller counters have been used. Fig. 1 shows the dipping-type counter, after Bale, Haven and Lefevre (*34*), as used with a pivoted beaker shelf and a 15-ml cell supported in a plastic holder. These counters are not self-quenching and operate at about 1,200 volts with a background of about ten counts per minute unshielded. Since they are usually light-sensitive, they should be enclosed in a light-tight box with a door arrangement for the placing of samples. The sensitive portion of the counter is a cylinder about 51 mm in length and 11 mm in diameter. As will be dis-

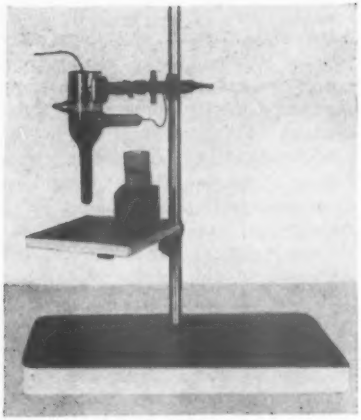

FIG. 1. Dipping-type Geiger-Müller tube with 15-ml cell supported in plastic holder on pivoted beaker shelf

cussed later, a test tube of 15-mm inside diameter and about 60 mm in length should be used with 2 ml of solution for maximum sensitivity. However, when the samples contain sufficient activity, it is more convenient to use 15 ml of solution in a cell of 22.4-mm inside diameter and 63 mm in length, as shown in the illustration.

No significant differences were observed when the cell was centered with the counter tube merely by inspection. Reproducible results were obtained with the counter tube and beaker shelf fixed in position and a given volume of solution in different cells of identical dimensions.

Since the activity comes in actual contact with the counter, it is essential that the counter be brought to background between measurements. When the samples measure below 1,000 counts per minute, this can be done simply by immersion in water or solvent and wiping with cloth. In a series of measurements, it is desirable to avoid infrequent highly active solutions, since it may then require considerable washing to reduce the count to background. In practice it is convenient to measure the highly active solutions with the cylindrical beta counter described below. It may be difficult to reduce the count to background where there is a tendency for the radioisotope to be adsorbed onto glass; it is valuable, however, to know that such adsorption is occurring, since losses can then be anticipated in the preparative chemical manipulations. Another type of thin-wall dipping counter, commercially available, has a diameter of 14 mm and a length of thin-wall section of 3 inches; this is a non-light-sensitive, self-quenching counter with reportedly good plateau characteristics.

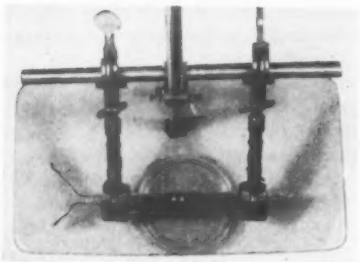

FIG. 2. Cylindrical thin glass wall beta counter as used to measure solution contained in standard Petri dish

Fig. 2 shows an arrangement employing a cylindrical thin-glass-wall beta counter with the solution contained in a standard Petri dish. This counter has a wall thickness of 30 mg/cm^2 and operates at about 900 volts with a background of about 35 counts per minute unshielded. As can be seen from the illustration, the geometrical efficiency of this arrangement is poor; nevertheless, when the samples contain sufficient activity, the convenience of this procedure has much to recommend it. This counter is considerably sturdier than the thin-mica-window counter and has much better plateau characteristics

than the dipping tube. Although for the purpose of the experiments described here it is less sensitive than the dipping counter, it does have the advantage of requiring no cleaning between measurements. Flat-bottomed Petri dishes which can be used interchangeably with good results are commercially available. Smaller dishes can be used to increase the geometrical efficiency.

Fig. 3 shows a thin-mica-window counter as used for measuring activity in solution. It may be obtained with a window thickness of 2.0 to 3.0 mg/cm^2 and operate at around 1,000 volts with a background of about 35 counts per minute unshielded. These counters have good plateau characteristics, but do not usually have a long life. With the type of emitters under discussion, the increased sensitivity of this counter will usually not compensate for its fragility as compared with the dipping and cylindrical glass wall counters.

Table 2 shows a comparison of the sensitivities obtainable for Cu^{64}, Mo^{99}, $Co^{56,57,58}$, P^{32} and I^{131} with the different measuring arrangements. The specific activity estimates are based on information furnished by the supplier of the isotopes. With the exception of the iodine values, the sensitivity data are reported as the actual micrograms of the element of the given specific activity required to give a count equivalent to the background count of the instrument. While it is recognized that this is an arbitrary method for an expression of sensitivity, the values should be of use in estimating requirements and dosages for the specific isotopes listed. In the case of iodine, since there was no carrier added to the preparation, the values are reported as the microrutherfords of activity required to give such an equivalent count. Because they were obtained from an intercomparison series furnished by the National Bureau of Standards, these activity values are considered

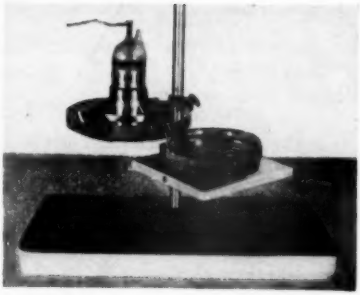

FIG. 3. Thin mica window counter as used to measure solution contained in standard Petri dish on pivoted beaker shelf

reliable. The necessity for expressing activities in three different units emphasizes the need for standardization of nomenclature.

The cells as used with the dipping counter have already been described. For the measurements with the other two counters, the thin film was prepared by allowing 0.05 or 0.1 ml of the isotope solution to be evaporated on glass over a circular area approximately 10 mm in diameter. The glass plate was placed as close as possible to the counter for the measurement. Since this area was slightly less than the diameter of the cylindrical counter and of the mica window, this procedure resulted in good geometrical efficiency as well as a minimum of self-absorption. The large Petri dishes were about 90 mm in diameter and 20 mm high. The small dish, which showed the effect of improving the geometrical efficiency without materially decreasing the self-absorption, was 25 mm in diameter and 12 mm high. The dishes were placed at the same distance from the counter for all measurements so that the greater the volume used, the closer was the surface to the counter tube.

A comparison of the sensitivity values between isotopes shows high sensitivity

TABLE 2

Sensitivity Values

(Micrograms of the Element Which Gave a Count Equivalent to the Background of the Measuring Instrument)

	Phosphorus	*Cobalt*	*Copper*	*Molybdenum*	*Iodine**
Item No. (from Table 1)	1	7	10	13	14
Estimated specific activity at time of measurement	4×10^4 mc/gm	1×10^4 mg radium gamma ray equivalent per gm	70 mc/gm	5 mc/gm	No carrier added
Dipping Counter (Fig. 1) Background 10 c/m					
2 ml Cell	1.9×10^{-6}	0.3×10^{-4}	0.5×10^{-2}	4×10^{-2}	19*
15 ml Cell	3.7×10^{-6}	0.4×10^{-4}	2.3×10^{-2}	22×10^{-2}	70
Thin Wall Beta Counter (Fig. 2) Background 40 c/m					
Thin Film	1.8×10^{-6}	0.4×10^{-4}	0.5×10^{-2}	2×10^{-2}	11
2 ml in Small Dish	12×10^{-6}	1.8×10^{-4}	14×10^{-2}	19×10^{-2}	298
5 ml in Large Dish	10×10^{-6}	2.8×10^{-4}	6×10^{-2}	13×10^{-2}	163
25 ml in Large Dish	23×10^{-6}	3.4×10^{-4}	22×10^{-2}	40×10^{-2}	630
50 ml in Large Dish	41×10^{-6}	3.5×10^{-4}	34×10^{-2}	79×10^{-2}	960
Thin Mica Window Counter (Fig. 3) Background 40 c/m					
Thin Film	1.6×10^{-6}	0.04×10^{-4}	0.1×10^{-2}	0.8×10^{-2}	3
2 ml in Small Dish	8×10^{-6}	0.8×10^{-4}	4×10^{-2}	10×10^{-2}	96
5 ml in Large Dish	18×10^{-6}	2.7×10^{-4}	4×10^{-2}	15×10^{-2}	122
25 ml in Large Dish	42×10^{-6}	4.0×10^{-4}	19×10^{-2}	47×10^{-2}	445
50 ml in Large Dish	78×10^{-6}	4.4×10^{-4}	34×10^{-2}	94×10^{-2}	850

* Iodine values expressed in terms of microrutherfords instead of micrograms

for those produced by a transmutation reaction, namely, cobalt, phosphorus and iodine, and relatively low values for copper and molybdenum, in which case the target element was chemically identical with the radioisotope.

In the case of phosphorus it may be noted that there was no significant gain in sensitivity when a thin film was used as compared with measurement by the 2-ml cell dipping counter. With the more convenient 15-ml cell, a decrease of only twofold in sensitivity was found. The mica-window counter offered no advantage over the cylindrical glass-wall counter. With cobalt, the thin-film mica window arrangement offered a tenfold increase over the other setups; here again the 15-ml cell was about as efficient as the 2-ml cell. When the Petri dish was used with either the mica-window or glass-wall counter,

there was approximately a tenfold decrease in sensitivity as compared with the dipping tube. With copper, molybdenum, and iodine, the thin-film mica window measurements were about five times as sensitive as the 2-ml dipping tube or thin-film glass wall counter measurements. Likewise, the 2-ml cell measurements were about fivefold more sensitive than those with the 15-ml cell. Where the Petri dishes were used with these isotopes, there were no great differences between the mica window or glass wall tubes. Some of the factors involved in the measurement of radioactivity in solutions have been discussed in a recent paper by Solomon and Estes (*35*).

In general, when the samples contain sufficient activity, the glass wall counter with the large Petri dish is the method of choice. If it is desired to increase the sensitivity, the dipping counter may then be used. With the isotopes under consideration here, there seems little need to employ the thin mica window counter except perhaps for ultimate sensitivity with thin films of those isotopes having a soft emission.

In regard to sensitivity, a very important consideration is the size of the sample available and the minimum volume of solution to which the sample can be made. Where only a small sample is available and this sample can be made to 1 or 2 ml, such as the small sample provided by the thyroid, pituitary or adrenals of the rat, the best procedure is to use the 2-ml volume with the dipping counter. If the sample could not be conveniently reduced to less than 1 ml, there would be no effective increase in sensitivity by making a thin film from 0.1 ml. In addition to the difficulties in the preparation of the film, there must be considered the inaccuracies introduced in working with small volumes. Where larger amounts of tissue are available, it has been most convenient to work with 20 to 50 gm samples and volumes of 25 to 50 ml. From the data in Table 2 and experience in handling of samples, it is possible to take into account such factors as sample size, volume of sample solution, and efficiency of measurement technique so as to provide for optimum conditions of assay for the specific experiment.

Preparation of Samples for Measurement

The body fluids such as blood, plasma, urine, bile or milk may be assayed directly if sufficient activity is present. If necessary, such samples up to 250 gm, can be ashed and concentrated to volumes of about 50 ml. It is desirable to reduce solid samples to a homogeneous liquid. The customary ashing procedures of analytical chemistry may be used; however, it is often possible to shorten them considerably since the requirements for radioactive assay are not as stringent as for chemical trace analysis.

When the conventional method of dry-ashing in the muffle furnace is used, care must be taken to avoid losses due to volatilization, incorporation of the isotope into solid carbon particles, or adsorption of the element to the crucible. In studies with short-lived Cu^{64}, this method of dry-ashing was found too time-consuming and a rapid method of pseudo-wet-ashing, based on solvent extraction of the fat, was developed so that 200 samples could be prepared for measurement within a few hours (*36*). The procedure follows: A representative sample, up to 50 gm, is cut into small pieces and placed in a 400-ml beaker, to which 40 ml concentrated nitric acid are added. The tissue is allowed to soak 10 minutes, after which gentle heat is applied. The resulting solution is evaporated to about 15 ml on a hot plate and then transferred to a steam bath for continued evaporation to remove the acid. The residue is washed into a separatory

funnel with small amounts of warm water. The beaker is rinsed with two 10-ml portions of iso-amyl alcohol which are in turn transferred to the separatory funnel. The separatory funnel is shaken gently and the two solutions separated and made to volume for measurement with a Geiger counter. The activities as measured in each solution are added to give the total for the sample. Iso-amyl alcohol was the most suitable general solvent tried, chiefly because of its high boiling point.

Certain tissues may require slight modifications of this procedure and the following suggestions are based on experience in this laboratory.

When it is necessary to pseudo-ash over 35 gm of whole blood, it has been found advantageous to slow down the initial reaction by using increased amounts of acid and applying heat very cautiously with frequent stirring. In the case of teeth, bone or cartilage samples, care must be taken not to reduce the acid solution to the point where the salts start to precipitate; it is very difficult to get the precipitated salts back into solution. In handling samples such as white bone marrow, lymph glands and adipose tissue, which contain large amounts of fat, there is a tendency for bumping and spattering so that it is advisable to heat them gently at all times, preferably on a water bath. With this type of sample, a solvent mixture of 10 parts ethyl alcohol, 25 parts diethyl ether and 25 parts petroleum ether may be more efficient than iso-amyl alcohol.

Some feces samples, depending on the nature of the diet, may require more than the usual amount of nitric acid for complete digestion. It has been observed that if these samples are evaporated to dryness, it is very difficult to get the residue into solution again. In some experiments, collections are made on filter paper and in such cases wet-ashing may be carried out as follows: The filter paper is first dampened with water and 4 to 5 ml concentrated sulfuric acid are added. About 10 ml nitric acid are then added, the beaker is heated gently until the violent reaction has subsided, and then heated further until the contents are charred. At that stage, 25 to 30 ml nitric acid are added and the digestion continued as previously described.

In some studies, it has been necessary to concentrate samples of milk for the activity measurements. A convenient procedure, combining the wet- and dry-ashing techniques, is as follows: 150 ml of milk is pipetted into a 400-ml beaker containing a few glass beads and 40 ml concentrated nitric acid is added. The mixture is heated gently until the foaming has subsided and then evaporated to reduce the volume. The solution is then heated on a steam bath until the excess nitric acid is removed. The solution is transferred to a 90-ml porcelain crucible using warm wash water and taking care that the total volume of the sample does not exceed 50 ml. The solution is dried at 100–105° C for about 12 hours and then ashed in a muffle furnace at about 500° C for a minimum of 5 hours. The ash is taken up in about 8 ml of an acid solution (100 parts of 2 N HCl and 20 parts concentrated HNO_3) and made to volume. If it is necessary to concentrate the milk more than 150 to 25, more than one 150 ml batch can be started and the final ash solutions combined.

Health Precautions

In using radioisotopes the necessity for adequate protection of personnel from radiation exposure must always be emphasized. There have been several excellent discussions of this subject recently (*37, 38, 39, 40, 41, 42, 43, 44*) and an authoritative report entitled, "Safe Handling of Radioisotopes," prepared for the National Committee on Radiation Protection, is expected to

be available in the near future.* A detailed discussion of Health Physics is beyond the scope of this review. However, since the extent and nature of precautionary measures are determined by the levels of radioactivity encountered and the specific types of experiments, it may be of interest to present a few of the simple techniques found adequate for the type of tracer study under consideration. It should be remarked that the health precautions at this level do not make the operations cumbersome or difficult.

The control of radiation exposure is directly dependent upon availability of adequate instrumentation. The following monitoring instruments are all commercially available. A battery operated portable Geiger-Müller counter, range from 0.05 to 20 mr per hour, has been found very useful in the survey of laboratory equipment, clothing and hands for low-level contamination. A portable ionization-type instrument, the "Cutie Pie," range 2 to 5,000 mr per hour, is more useful near and above the tolerance level. An advantage of the latter instrument is that it permits an estimate of allowable time of exposure should it be necessary to work momentarily with greater tolerance levels. A similar type of instrument known as "Zeus," which has a 1 to 2 liter ion chamber with a wire mesh window and two types of screens to allow discrimination between alpha, beta and gamma radiation, is also commercially available. The portable Lauritsen Electroscope, a relatively rugged quartz fiber instrument, is regarded highly for health protection monitoring, chiefly because of its retention of calibration and greater reliability; it is somewhat more sensitive than the "Cutie Pie." A fountain-pen-type ionization chamber is also useful for measurement of exposure of personnel dealing with sufficiently penetrating radiation. It is to be emphasized that these instruments should be calibrated frequently since battery or other failure may reduce their efficiency. This may be done by using a suitable radium button. For low-level detection of low-energy beta emitters, a count rate meter employing a thin mica window tube may be employed. It seems likely that in the future modified alpha counters may be convenient for this purpose. Film badges should be used to supply a permanent record of radiation exposure of personnel.

External exposure can be avoided by appropriate shielding or remote control operations. In tracer studies external exposure is usually important only in the handling of the original materials or in the preparation of the doses. The primary operation of putting the material into solution is a simple one and can be performed at distances of 2 to 6 feet by using readily improvised procedures; a magnetic stirring device has been found very useful for this purpose. Although not usually necessary, a surveying telescope can be used to read burettes at a distance. After establishing the procedure by practice with the inert element, and using equipment available in the ordinary laboratory, no difficulties have been encountered in handling up to 20 millicuries of copper, molybdenum, iodine or phosphorus as listed in Table 1.

There is always the problem of removing aliquots from the original solutions. Fig. 4 shows the withdrawal of solution from a test tube enclosed by two lead bricks. Holes are bored in both bricks to accommodate the test tube. In addition, a hole is provided in the upper brick just large enough to permit passage of a pipette or hypodermic needle. The cone of radi-

* A limited number of copies of the third draft of this document are available now from Dr. H. M. Parker, the National Committee on Radiation Protection, c/o Hanford Engineer Works, Richland, Wash., or from the AEC Isotopes Division, Oak Ridge, Tenn.

FIG. 4. Removal of radioactive solution, by means of syringe for dosage purposes, from stock solution enclosed in lead

ation passing through this hole can easily be avoided by working off center.

In the administration of radioisotopes to animals by injection, it has been found convenient to insert the needle itself with the fingers, and then to hold it in place with forceps while the syringe, held with tongs, is brought into position. The fingers need come no closer to the activity than 6 inches or so when the solution is injected. When necessary, the syringe can be shielded by lead. Similar procedures can be used for administering solutions by stomach tube. The adjustment of the dosage to the animal so as to have the samples contain the order of magnitude of activity necessary for Geiger counter assay is important in minimizing the radiation exposure of personnel as well as in reducing contamination of equipment. In a tissue distribution study, it is sometimes necessary to administer orally large amounts of activity because of poor absorption of the element. In such cases, if the animal is posted after a short time interval, care must be taken in handling the intestines, since considerable activity may be present in the contents. Adequate disposal must be arranged for highly active excretions and animal remains in addition to waste radioactive solutions.

It has been the policy in this laboratory that personnel should be exposed to no extraneous radioactivity whenever possible, and in no case to more than 0.1 of the maximum permissive tolerance of 12.5 mr per hour for any prolonged period. During several years, it has very seldom been necessary that workers in this laboratory be exposed to as much as 12.5 mr per hour, and then only for a few minutes at most.

Internal radiation effects are considered more hazardous than external exposure. Radioisotopes may enter the body by ingestion, inhalation or absorption through intact or injured body surface. The elimination of such entrance into the body is accomplished by observance of a "no smoking, no eating" rule in the laboratory, the use of surgical gloves to avoid skin contamination, the use of adequate hoods where the radioisotopes might be volatile, and the strict avoidance of dusting. The most dangerous isotopes are those which are readily absorbed into the body, show a selective accumulation in a small organ or bone, are only slowly eliminated from the body, and have a long half-life.

Considering the elements listed in Table 1, those most dangerous from the internal radiation standpoint are Ca^{45}, Fe^{55}, Mn^{54} and Zn^{65}; the less dangerous isotopes include Co^{60}, because of poor absorption, Fe^{59}, I^{131}, and P^{32}, because of their shorter half-lives; Cu^{64} and Mo^{99} are the least dangerous because of their short half-lives. Further information on the specific metabolism of these elements, which may be important in evaluation of the hazard, will be presented in a later paper in this series.

As far as the safe handling of radioisotopes is concerned, there can be no substitute for meticulous laboratory technique and an understanding of the

chemical, physical and metabolic behavior of the element, the nature of the radiation, and the performance of the monitoring instrumentation relative to the specific radiation involved.

The next paper in this series will deal with the specific nutritional trace elements from the standpoint of the existing problems, the radioisotope techniques which have been used, and the contribution of the radioisotope studies to the understanding of the system.

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STRAY-RADIATION MEASUREMENT WITH X-RAY FILM

X-ray film can be used to make quantitative measurements of small amounts of stray radiation. Here are measurement techniques and the necessary precautions to secure accurate and consistent results for exposures as small as 0.05 roentgen

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THE MEASUREMENT of low intensities of X-rays, gamma rays, and beta rays is necessary to ensure for personnel adequate protection from the harmful effects of stray radiations from radioactive materials and from electron and ion accelerators. Measurements may be made either to determine the intensity of a radiation in roentgens per unit time, or to determine the total number of roentgens received in a period of time. For this second type of measurement, X-ray film can be conveniently used.

This method of measuring exposure, while well known, has been used in the past as a relatively rough measurement of quantities of radiation several times larger than the daily tolerance. By using high-speed industrial-type X-ray film and by exercising care in all operations, it has been found possible to measure exposures as small as 0.05 roentgens with considerable consistency and accuracy and larger exposures with even greater accuracy. This discussion is concerned with the precautions and methods used in making accurate measurements of radiation with X-ray film, particularly necessary in measuring small quantities of radiation.

To measure radiation with film, the procedure, in outline, is as follows. From a number of pieces of film, alike in all respects, a few are chosen and to each of these a different exposure is given, the range of exposure being chosen to correspond to the range to be measured. After development of the films and measurement of their density, a calibration curve is plotted relating the density produced to the exposure given. The remaining pieces of film are used for measurement and the exposure, in roentgens, received by each is deduced from its measured density and from the calibration curve.

For this method to be reliable, it is essential that the pieces of film be alike initially and that they be given identical treatment except for the periods of exposure. The specific problems involved in satisfying these conditions and securing maximum sensitivity and reliability are dealt with below.

The information set down in the following sections relates primarily to the gamma rays from radium and to X-rays of corresponding energy; for other radiations additional data or suggestions are added where the information with regard to gamma rays is not relevant or is not adequate.

Choice of Film

The amount of radiation to be measured determines the choice of films. This is usually a rather small quantity and the fastest film available is required. Occasionally, however, measurements of large quantities of radiation must be made and for these

a slow film is more suitable.

An amount of 0.1 roentgen of radiation is frequently taken as the maximum amount to which any individual may be exposed in any one day; 0.1 roentgen per day is then called the tolerance dose and the radiation intensity which would give an exposure of 0.1 roentgen in eight hours is called the tolerance level. Thus a measurement commonly required is that of an exposure of the order of 0.1 roentgen.

The lower limit of exposure which can be measured with film is set by the smallest increase in blackening, above the background density of the unexposed film, which can be consistently produced and measured, and this amount is about 0.10 density units. For one of the fastest X-ray films available, Kodak Industrial X-ray Film "Type K," used with intensifying screens, this increase in density corresponds to an exposure of 0.03 r. Under the same conditions, for this film, an increase in density of 0.15 corresponds to an exposure of 0.05 r and an increase in density of 0.25 corresponds to an exposure of 0.08 r. This last amount can be measured with an accuracy of about 10% and 0.05 r can be measured with an accuracy of approximately 20%. In the slower speed, medical-type films, either screen or non-screen types, the exposure corresponding to each of the density increases mentioned above is about two or three times as great.

The greatest exposure which can be measured is limited by the greatest density which can be measured, usually about 3.0. For the fastest film this limit corresponds to 2.0 r. For the medical non-screen type of film, the limit corresponds to approximately 3.0 r, and for the medical screen-type film, it is a little larger, about 3.5 r. By using dental film, or the slow-speed, fine-grain, industrial type of film, a still greater maximum exposure can be measured. The exact maximum, minimum, and range, for any type, depends on the particular batch of film and on the developer and developing technique used.

For measurement of the smallest exposures, the high-speed industrial-type film should be used; for the largest exposures, the industrial fine-grain film should be chosen; for covering the greatest range of exposures, a medical screen-type film is best. Characteristic curves of the various types of film have been published (*1, 2, 3*) or may be obtained from the manufacturers.

The figures given above for exposure and corresponding density apply to gamma rays and to X-rays of energy greater than 250,000 volts; for X-rays of less energy, the figures will depend greatly on the wavelength of the X-rays, *i.e.*, on the kilovoltage and filtration used in producing them.

Transportation and Storage

Films are affected by a number of physical agents such as X-rays, gamma rays, beta rays, light, heat and humidity. In transportation and storage there may be opportunities for some of these agents to act on the films. If some exposure to them cannot be avoided, it should be kept as uniform as possible for all films within any one lot or box.

Any exposure to X-rays or gamma rays is recorded on the films and its effect varies from a slight increase in the background or basic fog to complete blackening of the film. A slight increase in the normal small-fog density does not harm a film for radiographic work, but it does decrease the sensitivity and accuracy which can be obtained in measurement of stray radiation with the film. Small exposures have been given to film by shipping radioactive materials in the same express car, by leaving an industrial radiographic radium capsule near

film in storage, by leaving a load of new film on a hand cart near a radium storage vault, by moving film past an inadequately shielded X-ray unit while it was in operation, or by storing film in a location where a very low intensity of X-rays and gamma rays acted on it for a long period. In some cases, because of the shielding of part of the film by the rest or by an opaque object, the increase in fog level varied from one box to another and from one part of a sheet of film to another.

If radiation is present in the area where the film must be stored, a lead safe of sufficient thickness should be provided to reduce the radiation intensity to the point where it will not affect the film appreciably during the maximum storage period.

Any exposure to beta rays with energy sufficient to penetrate the wrapping will also damage the film. It will expose it non-uniformly since beta-ray intensity falls off rapidly with increasing penetration.

The background-fog level on the film increases with the age of the film so that fresh film should always be used for measurement purposes. The supply on hand can be kept at a minimum by arranging with the supplier for delivery at about the rate of consumption.

The rate of increase of the background fog rises very rapidly with the temperature and, in this sense, film ages very quickly when stored in a hot room. Whenever possible, film should be kept in a cool place, preferably in refrigerated storage.

Although the effect of humidity alone on film is not certain, it has been found that under hot and humid conditions film stored in contact with lead intensifying screens for more than a few hours shows considerable irregular blackening.

Since the sensitivity of film is greater the lower the exposure it has received, and since non-uniformity of initial state destroys accuracy of measurement, it is important that storage and transportation be arranged to minimize any possible exposure and to keep unavoidable exposures uniform for all films.

Exposure

The type of packet used for dental X-ray film is very convenient since it provides either one or two small films individually wrapped in a light-tight paper package. The type of film used in dental X-ray packets is much too slow for most measurement purposes, but other types, including Kodak "Type K," can be secured on special order with the same packaging. The film packets are about 1.2 by 1.6 inches, by $\frac{1}{32}$ inch thick, and plastic or metal badges to hold one or more such films are easily devised.

Because of the additional correspondence, delay and cost, involved in obtaining high-speed films in a dental-type package, it is often better to use readily available, standard, radiographic sizes of X-ray film, and to cut and wrap individually the small pieces used for measurement. Light-tight paper, obtainable from the film companies, can be folded and sealed to make suitable envelopes, or special light-tight film holders can be devised.

A design of holder for a small piece of film is illustrated in Fig 1. A piece of film is held between two thin circular aluminum foils, which are pressed together at the circumference by two inter-penetrating magnesium rings. The two rings are so designed that when screwed together they form a light-tight trap and make a light-tight seal by clamping the aluminum foils between their inner circumferences. Lead-foil intensifying screens, which increase the blackening of the film for a given exposure, may be placed on each side of the film, next to it.

In some measurements it is desirable to get a rough idea of the quality of the

radiation being measured. One means of doing this is to use an absorber such as a piece of lead sheet, 0.020 inch thick, between the film and source of radiation. By comparing the density of the film in the area under the absorber with that in the area adjacent to it, the presence of soft, easily absorbed radiation may be detected. A more quantitative device is a step penetramometer made up of a series of thin foils of copper, lead or aluminum, covering a part of the film. Still another means is the use of lead intensifying screens adjacent to the film, with holes in the front and back screen so that, from the densities in the areas exposed by these holes as compared with the density of the rest of the film, some deductions can be made about the incident radiation. These devices can be utilized in some cases, particularly where the film is fixed in place during exposure, to determine the predominant direction of incident radiation.

In the film holder illustrated, each lead-foil intensifying screen has a circular hole in it. The hole is centered in the front screen and is off-center in the back screen, the two overlapping by about one third of their diameter. Four areas are thus provided on the film; an area with front screen only, one with back screen only, an area with both front and back screen and an area with neither front nor back screen. The density produced in the area with no front or back screen is a measure of the exposure. If the radiation is predominantly beta radiation and soft gamma radiation from the front, the areas protected by the front foil are much less blackened than the areas with no front screen; the area with the back screen only is as dark as, or darker than, the area with neither front or back screen. If the radiation is predominantly hard gamma radiation, the area with both front and back screens tends to be slightly darker than the area with no screens, since absorption in the front screen is small and the intensifying action of front and back screens is appreciable. By comparing the density patterns of the four areas with those produced by the calibrating radiation, or with other known radiations, considerable information can be secured.

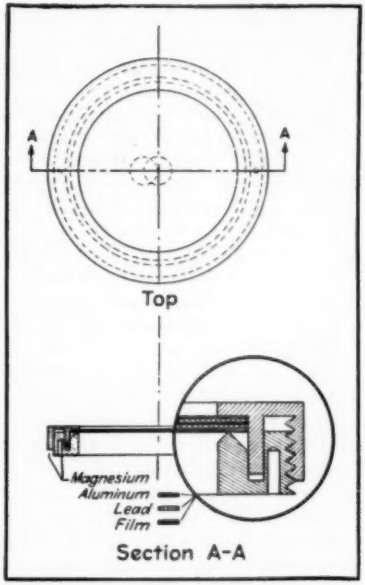

FIG. 1. Exposure holder for cut film

Measurement films are usually attached at waist or chest level, *i.e.*, close to the center of the body mass. In some surveys films have been worn at the same time on the chest, at the waist, at the knee, and on the wrist. The genetic effect of radiation is the effect about which most concern is felt, so that films are best worn at waist level, unless the nature of the work or the type of shielding provided is such that large exposures may reach other parts of the body without affecting the waist level film. For some kinds of work a film

attached to a watch strap on the right wrist is used and special rings have even been provided to carry small bits of film on the hand.

In plants where radioactive materials are handled, films are frequently fixed in various locations to secure information on the average radiation-intensity levels. By using lead protection on one side of the film, information on the directional intensity of the radiation can be secured and, by using the devices mentioned previously, information on the quality of the radiation can be secured.

Two films of the same kind are sometimes placed in a single packet or holder in order to get a greater total change in density in a given time. Two films of different speeds, such as Kodak "Type K" and "Type F," one fast and the other slow, have been used in the same holder to get the maximum latitude in measurement, so that any exposure, from very small one to very large one, can be measured. If this is done, the film types should always have the same relative position in the holders in measurement and in calibration; the fast film is usually placed at the front.

Identification of the films must be very carefully done. A number is given the loaded film holder and transferred to the film when the holder is opened for development, or the number is put on the film when it is loaded and is placed at the same time on the film holder. Numbers written on the film with a hard pencil and firm pressure are easily seen after the film has been developed; a film-numbering perforator may be used if the numbers do not obscure too much of the film.

Calibration

Films of any one emulsion type are not perfectly uniform. The emulsion is prepared and put on the film base in batches; therefore, the characteristics of the finished film may vary slightly from batch to batch and there may even be small variations within a batch. Though these differences in characteristics and in initial condition are small, errors due to them are avoided by treating a box of films, or part of a box of films, where a large box is used over a period of time, as a unit and using a separate set of calibration and control films for each unit.

In the case of dental-type packages, a calibration is prepared for each box of 72 films. As soon as the box is opened, all films are numbered and six films, one from each dozen, are selected for the calibration from which the exposures to be measured with the remaining films will be evaluated. Two control films, one from each end of the box, are taken at the same time to be developed with the calibration films. Four other control films are set aside as the measurement films are used, at the rate of one control film for each fifteen measurement films. Control films are given no exposure but accompany the calibration and measurement films at all times except for the period of intentional exposure. They are developed along with the calibration films, or with the particular group of measurement films to which they belong, and provide a measure of inherent background, of any stray radiation, and of the processing technique.

Where small films cut from larger sheets are used, a group of 80 to 100 films, cut from adjacent sheets in a box, is taken as the unit for calibration, and the calibration and control films are selected from this group, distributed through it, in a manner similar to that described above. The number of calibration and control films may be varied to suit the circumstances; it has been found better to use the rather high proportion suggested above than to use fewer films and to have occasional doubts about the accuracy of the calibration or the uniformity of conditions.

For radium gamma rays, the calibration can be carried out by placing the six films at different distances from a radium capsule for a fixed time or at a fixed distance for varying times, or by using any other combination of time and distance factors. The appropriate times and distances can be calculated using the relationship that 1 mg of radium in a 0.5 mm platinum shield will produce 8.4 r at 1 cm in 1 hour, or 0.14 r at 1 cm in 1 minute. In equation form

$$r = \frac{t \times m \times 0.14}{d^2}$$

where r = roentgens received by film
t = time of exposure in minutes
m = mg of radium in capsule
d = distance from radium to film in centimeters.

The number of milligrams of radium is known and a set of calibration exposures, covering the range to be measured, can be chosen. For each such exposure, the time or distance can be found by choosing a suitable value for the other.

For measurement of exposures near the tolerance amount, a suitable set of calibration exposures is: 0.05 r, 0.10 r, 0.20 r, 0.40 r, 0.80 r, 1.60 r. If greater accuracy in the lower part of the range is sought, two of each of the two lowest exposures may be made, and the 0.80 r and 1.60 r exposures may be omitted. It is best to use a fixed distance and a varying time since all films then receive the same contribution of radiation scattered from the source and from the air. Distances of less than 25 cm are not used. The films are so supported that there is nothing behind or adjacent to them, or to the radium source, from which radiation could be scattered. Wooden stands supporting films and radium six inches above a wooden table top have been found to be satisfactory. For convenience, the times corresponding to a set of calibration exposures at two distances for 300 mg of radium are given in the table above.

	Time in Minutes	
Roentgens	*Distance = 29 cm*	*Distance = 58 cm*
0.05	1.0	4.0
0.10	2.0	8.0
0.20	4.0	16.0
0.40	8.0	32.0
0.80	16.0	64.0
1.60	32.0	128.0

A typical calibration graph is reproduced in Fig. 2.

The calibration prepared for gamma rays cannot be used to evaluate the exposures produced on films by X-rays generated at less than 250,000 volts, since the effect on the film of a given number of roentgens varies with the wavelength of the radiation below this limit. For X-rays a calibration must be prepared by finding the film blackening which corresponds to each of a number of exposures as measured in roentgens by an air ionization chamber. If a sensitive calibrated ionization instrument is available, such as a Victoreen minometer, each piece of film can be exposed to the X-ray beam beside an ionization chamber, and the exposure received thus determined by measurement. If an ionization instrument of much less sensitivity is avail-

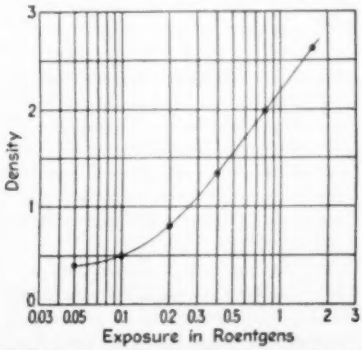

FIG. 2. Typical calibration graph. Kodak Type K film used with gamma rays from radium

able, such as a Victoreen r-meter with a 25-r chamber, it can be used to measure the exposure produced by a weak X-ray beam in a long time, and the calibration films can then be given short exposures, calculated to correspond to the chosen calibration exposures.

The quality of the calibrating radiation must be the same as that of the radiation being measured. Where measurements are to be made of a radiation that is mixed, the results will be on the safe side, since hard radiation is less effective in blackening film than soft, if the calibration is made with the hardest radiation to which the films are exposed.

If the stray radiation to be measured has been scattered and rescattered several times, a rather rough calibration can be made by arranging to scatter the primary beam one or more times before it reaches the ionization chamber and calibration films. On the other hand, if the radiation to be measured is that which has penetrated a protective wall and thus been filtered, the calibration can be made by arranging equal or greater absorbing material around the films and the ionization chamber so that only heavily filtered radiation can reach them.

If, for use with low-voltage X-rays, 80–100 kv, lead foil or tin foil 0.003–0.005 inch thick is substituted in the film holder for the 0.020-inch thick lead intensifying screens suggested earlier, the presence of a high percentage of soft radiation is indicated by the greater blackening of the uncovered areas. Calibration for the covered and uncovered area can be made by using the ionization chamber with and without coverings of the same foil.

For beta rays, a calibration cannot be made readily in terms of roentgens but can be made in terms of a beta-ray standard if one is available. Two recent articles give information on the measurement of beta-ray intensity in roentgens (*4*, *5*), and the presence or absence of beta radiation can be easily observed.

In making a calibration in this way it is assumed that the reciprocity law holds: that the blackening of the film is the same for equal values of the product of exposure time and intensity of radiation; *i.e.*, an intensity of 2 units for 1000 minutes has the same effect on the film as an intensity of 200 units for 10 minutes, or 1000 units for 2 minutes. This assumption is valid, or very nearly so, as long as the films are exposed with no screens or with metal intensifying screens but is not valid if chemical intensifying screens are used. Thus with intensifying screens or with lead screens, it is possible to produce a calibration curve, rapidly, with relatively high radiation intensities, and interpret effects produced with much lower intensities over a longer exposure period.

Development

In development, as in cutting films and loading film holders, the darkroom conditions must be maintained as nearly ideal and as uniform as possible, since small changes in darkroom conditions or techniques may have a considerable effect on the films.

It should be remembered that the safelights usually used in an X-ray darkroom do produce an exposure on film; the exposure produced does not harm film when used for radiographic purposes but it should be avoided on film used for measurement. The intensity of light on the working surface is normally greater than is needed after the eyes are dark-adapted and can be reduced by raising the safelight or by replacing the bulb recommended for it by one of smaller wattage. The intensity of light on the table top should not be reduced to the point where the speed of carrying out operations with the film is lowered. An area shielded from illumination by the safelight and

adjacent to the working space, where unprotected film can be left momentarily while other films are being manipulated under the light, is very convenient. By bringing the film under the safelight only for the few moments when it must be seen to work with it, and by carrying out as many operations as possible in the shielded area, the total exposure to the safelight can be made a fraction of that usually given. By careful planning of operations, arrangement of supplies, and adjustment of lighting intensity, a minimum of exposure can be given to the film and that exposure which is unavoidable can be kept uniform from one film to another.

Uniformity of development is very essential. A number of precautions are therefore necessary. Any standard X-ray developer may be used, but the quantity of developer must be large enough that its characteristics are not changed appreciably during development of the 60 to 100 pieces of film belonging to one calibration. A record of the number of films processed or, if the films vary in size, of the number of square inches of film processed, should be kept so that either a replenisher can be added or the developer replaced systematically. Replenishment or replacement of the developer is done only on completion of development of all the films dependent on one calibration.

Before starting development, the developer is stirred thoroughly so that it is at a uniform temperature, and is warmed or cooled so that its temperature is 68° F. Although compensation for temperatures differing from 68° F, within the range of 65° F to 70° F, can be made by varying the time of development,* this should be avoided if possible. In all cases, the calibration, control, and measurement films of any one group must be developed at the same temperature and for the same time. The time of development must be kept exact; 5 minutes at 68° F is the usual period, though for non-screen type films 8 minutes gives more complete development and greater density.

For uniformity of development, agitation of the film is necessary and the method of horizontal agitation described below has been found to be very satisfactory. On being put in the developer, the film clips are rapped sharply against the tank to dislodge any air bubbles and are then left for one minute. At the end of this time the left-hand holder is grasped firmly to keep it vertical in the solution, and moved slowly to the left-hand side of the tank. This process is repeated with each of the other holders till all are at the left. Then, in the same way, the right-hand holder is moved slowly back across to the right-hand side of the tank, followed in turn by the others. This moving of the films from one side to the other is kept up until three complete cycles—to the left, to the right, and to the original position—have been carried out, and the film holders are back in the original places. The films are pushed through the solution in the same way that a board held vertically may be pushed through water; the current across the front and the turbulence behind clear away the products of development from the surface of the film and bring fresh developer into contact with it. The whole process is repeated at the end of the third minute and if 8-minute development is used, at the end of the fifth minute.

At the end of the development period, the films are lifted from the developer, allowed to drain only for a second or two, and immersed in an acetic-acid stop bath. They are agitated in this bath for about thirty seconds and then transferred to the fixing solution. If the stop bath is not used, a 2-minute wash in constantly running water, with

* See manufacturers' recommendations.

agitation, is substituted. The films are transferred to the fixer, which is at about room temperature.

The fixing period is at least three times the time required for each film to clear, with a minimum of 10 minutes; it should be of fixed duration for any one group of measurement and calibration films. After fixing, the films are washed in running water, not colder than 50° F nor warmer than 70° F, for 20 to 40 minutes.

The films are dried in air at room temperature or higher, with or without forced draught; all from any one group are given the same treatment.

Measurement of Exposure

The exposure received by the measurement films is determined by comparing them with the calibration films which have had known exposures. This comparison can be made by eye, or by using some form of densitometer. When it is necessary only to ascertain that the exposure received by the measurement is less than a definite limit represented by a particular calibration film, or where an estimate of exposure is sufficiently accurate, the visual comparison is quite adequate and can be very rapidly made. If an accurate determination of the exposure received is required, the use of the densitometer is more satisfactory. By measuring the densities of the calibration films and plotting them against the exposures given, a graphical relationship can be obtained from which the densities of the measurement films can be translated into exposures received by them. A calibration of this type is illustrated in Fig. 2.

The photoelectric type of densitometer, though no more accurate than the visual-matching type of instrument, is more rapid and less of a strain on the observer. Ansco-Sweet and Zeiss densitometers, of the photoelectric type, and Eastman and Marshall densitometers, of the visual-matching type, have been found satisfactory. Others are available or can be improvised.

The different instruments do not agree very well in their density figures since the different optical systems measure different proportions of specular and diffuse transmission and may have different spectral responses. This is not important as long as calibration, control, and measurement films of any one group are compared with the same instrument.

For very dense films, blackened beyond the upper limit of the densitometer, which is usually about 3.0, the emulsion can be stripped off overlapping, but not coincidental, areas on opposite sides of the film and the density of each emulsion and film base can be read separately. From these figures total density can be deduced.

The background or fog density is about 0.20 to 0.40 for the fastest type of film and is about 0.15 to 0.25 for other types of film.

* * *

The methods and precautions described here were worked out at the National Research Laboratories and were applied later to study of radiation hazards involved in measurement of large quantities of radium at the National Research Laboratories, and in the refining of radium by Eldorado Mining and Refining (1944) Ltd., at Port Hope, Ontario. The assistance of Miss D. M. Driscoll in developing and measuring the hundreds of films used is gratefully acknowledged, as is the cooperation given by Mr. A. H. Ross, manager of the radium refinery.

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Measurement of Radioactive Isotopes

Recognizing the need for radioisotope comparison standards for precision measurements, the Bureau of Standards has developed such standards. Their preparation and use are outlined

THE DETERMINATION of the number of radioactive atoms in a sample of a radioactive isotope has become a matter of importance since such isotopes have not only become an article of commerce but are also used in biological and medical experiments and even in clinical treatment of diseases. Uniform quantitative results in all laboratories can be obtained only by using procedures that will yield absolute measurements or by the use of uniform standards of comparison that will, under proper conditions of measurement, give the same result whenever the determination is made.

The National Bureau of Standards has therefore issued standard samples of radioisotopes whose radioactivity has been accurately determined, and is planning to issue samples of additional radioisotopes as they become available.

Two factors are involved in the preparation and use of these samples. First, methods and standards that are correct in principle must be used. Second, a reasonable degree of accuracy must be attained in all steps of the process not only of producing but also of using the standards. In order that the biological, medical, educational, and industrial laboratories to which the standard samples of radioisotopes have been issued may use them in a way that will assure correct and reliable results, comprehensive and accurate data have been made available.*

Essentially the measurement of a sample of a radioisotope is the measurement of the number of radioactive atoms present. This determines the strength of the source and involves three basic concepts: (1) the total number of radioactive atoms present, N; (2) the disintegration, or decay, constant, usually represented by λ, and obtained from the half-value period, T, by the relation

$$\lambda = \frac{\ln 2}{T} = \frac{0.693}{T}$$

and (3) the disintegration rate, or activity, λN, the number of atoms disintegrating in unit time.

Measurement of the rate of disintegration is necessary in order to determine the total number of atoms present. The disintegration rate is, of course, equal to the number of nuclear particles, usually beta particles or positrons, which are emitted per unit time from all atoms which disintegrate within this interval of time. This is true since, in general, one of these particles is omitted for each disintegration which takes place. Important exceptions to this general rule will be discussed later.

The requirement that all disintegration particles must be counted per unit time is by no means simple. The principal difficulty is that these particles are emitted equally in all directions so that it is practically impossible to devise detecting equipment which will record all of them. The Geiger-Müller beta-ray end-window counter, for example, has a window through which the beta particles enter from a source placed in front of

* See National Bureau of Standards Circular C473 by L. F. Curtiss, Chief of the Radioactivity Section. This is expected to be available shortly from the Government Printing Office.

the window. The counter can record only that fraction of the particles which passes through it. This represents, from geometrical considerations alone, not more than 25% of the particles actually emitted. At least three other factors may distort the results. They are absorption of particles in the window; absorption of particles in the source; and backward scattering of particles by the support of the source which directs through the window some particles originally emitted in other directions. The first two factors reduce the number from the correct value and the third increases it.

Conceivably the contribution of all of these factors can be evaluated experimentally for a particular source and a definite arrangement of the apparatus. This not only requires considerable work but it must be repeated whenever changes are made in either the source or the apparatus.

Radioactive Standards

Most of the difficulties in making quantitative determinations of radioisotopes can be eliminated if standard sources of the radioisotope under measurement are available. A standard source consists of a preparation of the isotope in a form convenient for use with the detector of radiation, and for which the disintegration rate is known from previous calibration.

Obviously, to be useful as a standard, a radioisotope must have a relatively long half-life. Furthermore, of those which have a sufficiently long half-life only those which can be calibrated in absolute disintegration rates are acceptable for preparation of standards. The number of isotopes which satisfy both these requirements is, at present, very limited.

When the standard is prepared from the same isotope as that to be measured only three simple precautions are required to secure reliable results: (1) Readings must be made with the standard in the same position as that at which readings are made on the sample; (2) the sample must be uniformly distributed over approximately the same geometrical area as the standard; and (3) the sample must be supported on a layer of material identical with that supporting the standard, or at least one that produces the same back-scattering effect.

FIG. 1. Samples of radioisotopes are compared with the National Bureau of Standards RaD + E beta-ray standard (disk on base of stand) using a bell-type beta-ray counter (on stand) and recording instrument. In order to correlate measurements of radioisotopes in different laboratories, identical 2½-ml samples I^{131} were distributed to approximately 40 hospitals, universities, and similar institutions using this isotope or interested in its measurement. The range of values reported by these laboratories, which varied as much as 80 percent from the average, has been plotted on the graph at the right

Isotopes which have short half-lives and a known disintegration scheme can be measured with a beta-ray standard of some other isotope with fair accuracy if it is known that a beta ray or a positron is emitted for every disintegration, and if the maximum energy of the beta-ray spectrum of the standard is not too different from that of the beta-ray spectrum of the isotope to be measured. In comparing samples of isotopes differing from the standards, all precautions outlined for the comparisons of the same isotope must be employed and the

FIG. 2. Two gamma-ray standards of Co^{60} prepared and certified by the National Bureau of Standards to contain specific radioactive strengths—one ten times the other. The strength of these standard samples, issued in sealed 5-ml ampoules, is measured in rutherfords (rd). Such radioactive samples are used for calibration of unknown solutions of Co^{60}

thickness of the counter window in mg per cm^2 must be known. In addition, an absorption curve for the beta rays from the standard and a similar absorption curve for the beta rays from the sample must be taken. This procedure consists of observing the counting rates for each when different known thicknesses of aluminum in mg per cm^2 are interposed between the source and the counter. When the data are plotted on semi-logarithmic paper, with counts per second on the logarithmic y-axis and thickness on the linear x-axis, the graphs will be very nearly straight lines. Extrapolating to zero thickness gives zero absorber counting rates for both standard and unknown sample, from which the disintegration rate of the unknown sample can be computed.

Since calibrated standards supplied by the National Bureau of Standards can be expected to maintain their calibration only when handled carefully by trained personnel, it is desirable that laboratories prepare their own working standards. This applies particularly to beta-ray standards deposited on metal planchets, such as the RaD + E standards. There is another advantage in the preparation of working standards in the laboratory. This permits the primary standards in many instances to be prepared as gamma-ray standards consisting of a solution sealed in a glass ampoule from which it need never be removed for use.

An example is furnished by the Co^{60} standards now available from the National Bureau of Standards. They consist of 5 ml of solution containing a total of 1.5 rutherfords (1.5×10^6 disintegrations per second) in one series, and 0.15 rutherfords in another. They may be used to calibrate unknown solutions of Co^{60} by comparing the gamma-ray activity of the unknown solution in a similar ampoule, using a gamma-ray electroscope for the purpose. The ratio of these readings, corrected for background, provides the information for computing the strength of the unknown solution in terms of a disintegration rate.

After the unknown sample has been calibrated, it can be made up to a standard volume, diluted in known ratios, and aliquots taken to produce deposits of appropriate activity for beta-ray standards.

Unknown Disintegration Schemes

It has been shown that, without a knowledge of the disintegration scheme, it is impossible to make reliable measurements of radioisotopes in terms of disintegration rates. The disintegration scheme serves to determine whether the mode of disintegration is such that disintegration rates can be obtained from observations which can be made conveniently. For example, if the isotope is a positron emitter but also disintegrates in part by electron capture, there is no convenient way of meas-

uring the disintegration rate since it is difficult to determine the number of disintegrations occurring by electron capture. This capture can be detected only by virtue of the resulting characteristic X-radiation.

In the case of those isotopes for which disintegration schemes are lacking or for which it is known that the mode of disintegration does not lend itself to measurement of disintegration rates, an alternative method of comparison of activities of sources is available if the isotope emits gamma rays. This alternative does not give disintegration rates but it can, when properly applied, yield reliable comparisons of sources in various laboratories. For a particular isotope which emits gamma rays it is obvious that the intensity of the gamma radiation emitted is proportional to the amount of the isotope present. However, it is a well-known fact that electroscopes and ionization chambers used for measuring gamma rays have sensitivities which vary greatly from each other due to variations in size, materials, geometrical disposition of the source, and similar factors. Therefore, the comparison of source strengths by the gamma-ray method is a valid method only when confined to the same isotope and when using the same electroscope in the same geometrical relation to the source. This is the basic principle of all measurements of radium by the gamma-ray method.

A method has been proposed for extension of the gamma-ray method of comparison of sources to all laboratories. To obtain uniform results with such a method, no matter where the comparisons are made, a standard instrument and standard geometry are necessary. The requirement for the standard instrument is that it shall yield the same response for two equal samples of any isotope, regardless of the energies of the gamma rays emitted by these isotopes. There is a unit of gamma radiation which is defined without reference to the energy of the gamma ray. This is the roentgen, defined as "that quantity of roentgen or gamma radiation such that the associated corpuscular emission (secondary beta radiation) per 0.001293 gm of air, produces, in air, ions carrying 1 electrostatic unit of electricity of either sign." Therefore, an ionization instrument properly designed to measure roentgens will satisfy the requirement of a standard instrument. To determine the strength of a radioactive source the roentgens per unit time must be measured at a standard distance.

These considerations have led to the suggestion* that the unit of time be one hour and the distance one meter. Convenient magnitudes are thus provided for sources in common use. Furthermore, the distance of the source from the measuring instrument is relatively large and therefore small accidental variations in this distance will not produce serious errors, because the response of the instrument varies inversely as the square of the distance from the source. With this experimental arrangement the unit which is actually being used for comparison of gamma-ray sources is defined as 1 roentgen per hour at a distance of 1 meter (roentgen per hour at a meter, abbreviated rhm).

Units

This leads to a consideration of units for expression of disintegration rates. In 1910, at the Radiology Congress in Brussels, the name *curie* was applied to the amount of radon in equilibrium with one gram of pure radium. In 1930 the International Radium Standards Commission extended this definition to include the equilibrium quantity of any decay product of radium, with the comment "The Commission does not favor

* E. U. Condon, L. F. Curtiss, *Phys. Rev.* **69**, 672 (1946).

its extension to members outside the Ra family."* However, the use of the curie in referring indiscriminately to disintegration rates has occurred in numerous places in the literature on radioactivity. As originally defined and as amended in 1930, the curie refers to the rate of disintegration of 1 gram of radium since 1 curie of radon, by virtue of its definition as the quantity in equilibrium with one gram of radium, is also that amount of radon (or any other member of the radium family), which has a rate of disintegration equal to one gram of radium.

Consequently the curie can be applied only when it is intended to refer to rate of disintegration. Although the rate of disintegration of 1 gram of radium has never been measured with great accuracy, and the exact value is in dispute to the order of 3 or 4%, the National Bureau of Standards has adopted the arbitrary value of 3.700×10^{10} disintegrations per second for the curie when applied to isotopes other than members of the radium family.

For many purposes a unit for the disintegration rate can be used which is smaller than the curie and which can be specified exactly and independently of any natural constants, such as the rate of decay of radium. A convenient quantity for a unit is that quantity of radioisotope which disintegrates at the rate of a million disintegrations per second. The name *rutherford* (rd)† has been suggested for this unit. Features which make this unit simple to use include a numerical magnitude that is easy to remember, and a size that is frequently used in the laboratory. For example, a therapeutic dose of many isotopes will be of the order of 100 rd, beta-ray sources for use with mica-window, bell-type conuters will be of the order of 100 to 500 μrd, and the weakest source that can be measured with any accuracy with these counters is of the order of 1 μrd. Tracer samples will usually be of the order of 1 rd.

The use of the rutherford in data presupposes that a disintegration rate has been measured and that this rate is expressed in terms of disintegrations per second. This procedure, if rigidly followed, removes all confusion regarding units and renders data reported from different laboratories directly comparable on an absolute basis.

* *Revs. Modern Phys.* **3**, 432 (1931).

† E. U. Condon, L. F. Curtiss, *Phys. Rev.* **69** 672 (1946).

"Before the practical and useful applications of atomic energy as a source of power can be achieved, the engineering profession and the industry of this country must take an active part in its development, by acquiring the necessary knowledge in the atomic sciences as well as by acquiring practical experience in the atomic-energy field. This is essential, because the development of atomic power is predominantly an engineering problem, rather than a problem of new scientific discovery. As far as basic scientific information is concerned, more is probably known today about the fission of uranium or, say, the interaction of gamma rays with matter than is known about the basic mechanisms of combustion or supersonic aerodynamics.

—Andrew Kalitinsky, in a speech before the Society of Automotive Engineers, on June 16, 1948

HEALTH PHYSICS*

A brief summary of the principal phases of radiation health protection, dealing with the biological effects of radioactivity. The discussion embraces a consideration of permissible levels of exposure and the role of the health physicist

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SOON AFTER X-RAYS were discovered and concentrated radioactive materials were available, it was observed that high-energy radiations could produce serious damage to the human body. The gross effects of radiation absorption became known from animal experiments and a number of disastrous human over-exposures. A series of national and international conferences resulted in the establishment of provisional permissible exposure levels. Safety precautions based on these levels proved to be reasonably satisfactory when relatively small intensities were the rule and when the persons subject to exposure were few and specially trained.

The Health Physicist

When the all-out attempt to develop the atomic bomb was inaugurated, it was recognized that radiation hazards of unprecedented magnitudes would be encountered. Furthermore, many of the workers would be ignorant of these hazards and methods of avoiding them. To protect these workers against radiation injury, the profession of *health physics* was created.

The health physicist must have a thorough understanding of the physics of nuclear radiations and their effects on the human body. He must bridge the gap between physics and medicine and be able to cope with problems in waste disposal, ventilation and decontamination. Thus, it is evident that a health physicist requires broad, yet thorough, training in a variety of fields.

The past records of the radiation health protection groups are excellent. Even with the urgency of wartime and the use of large quantities of new radioactive materials, serious injuries were restricted to a few accidents due to violations of safety rules by trained personnel. It has been shown that almost any experimental procedure can be carried out with any amount of active material if proper precautions are taken.

With the increased availability of radioactive materials, the number of persons potentially exposed to dangerous radiations has increased many fold. Most of these workers appreciate in a vague way that the radiations are dangerous but too many have the "it can't happen to me" attitude. Unfortunately, the biological effects of the absorption of nuclear radiations are not as immediately apparent as a burn or a cut, and irreparable damage may be done before the victim is aware of it.

* Adapted from Chapter 18, "Nuclear Radiation Physics," by R. E. Lapp and H. L. Andrews (Prentice-Hall, New York, 1948).

This article is not intended to frighten workers away from the use of these most important substances. Rather it aims to describe the hazards involved, the methods of detecting and measuring the radiations, and some of the problems faced by the health physicist in insuring a safe working environment.

Radiation Hazards

There are at least seven forms of high-energy radiations which are potentially dangerous, but only three of these are commonly encountered in ordinary laboratory uses of isotopes. The others are encountered only in the vicinity of high-voltage particle accelerators, nuclear reactors, or atomic bomb detonations and will not be discussed here.

Alpha particles. Alpha particles, which are doubly ionized helium atoms, are ejected from radioactive nuclei with energies ranging up to about 10 Mev. They produce dense ionization as they pass through matter and consequently lose energy rapidly. Practically all alpha particles have a range of less than 10 cm in air and less than 100 microns in tissue. Because of their short range, alpha particles constitute a health hazard only when they are introduced into the body. Even if alpha emitters are closely adjacent to the skin, alpha particles will be entirely stopped in the cornified epithelium and will produce no injury to living tissue. Alpha emitters are, therefore, exclusively internal hazards.

Beta particles. Beta particles are high-speed electrons ejected from nuclei with initial energies ranging from 0 to about 5 Mev. They produce rather sparse ionization in traversing matter and consequently have ranges up to several meters in air and about 3 cm in tissue. Beta particles constitute an external hazard since they can penetrate significant distances into tissue from external sources. Beta emitters which have gained entrance into the body constitute a serious internal hazard, since practically all of the beta particles will be absorbed by the tissue.

Gamma rays. These are short-wavelength electromagnetic radiations emitted with the velocity of light from radioactive nuclei. They are usually emitted concomitantly with beta or alpha particles but in some cases only gamma radiation is emitted. Gamma rays and X-rays have identical properties and differ only in the method of production. Electromagnetic radiation does not have a definite range but decreases exponentially in intensity as it passes through matter. Gamma rays from strong sources can be detected at a distance of one hundred meters or more. They are obviously both internal and external hazards.

Absorption by Tissue

Alpha and beta particles are electrically charged particles and are directly ionizing. As a charged particle moves close to an electrically neutral atom the electric field of the particle will sweep over the atom and may pull an electron from it. The result is an ion pair.

Electromagnetic radiation is not directly ionizing but interacts with matter to produce high-speed electrons which are ionizing. Gamma rays may produce these high-speed electrons by any one of three different mechanisms, but from the biological standpoint the end result is the same: the conversion of the absorbed energy into tissue ionization.

Careful measurements indicate that an energy of about 32.5 ev must be expended for the production of one ion pair. Thus a 5.3 Mev alpha particle from polonium will produce, in tissue, $5.3 \times 10^6/32.5 = 163{,}000$ ion pairs in a distance of perhaps 100 microns. A beta particle of the same energy will produce the same number of ions over a

path of about 3 cm. The ionization density is not uniform, being greatest near the end of the path just before the particle loses its ability to ionize.

Units

A physicist would characterize a beam of radiation by giving the number of particles passing a reference point per second together with the velocity distribution, or he might specify the total energy flux per second. Such descriptions do not fit the needs of the health physicist for the biological effects depend upon the energy absorbed in the tissue and not on the amount that passes through. For example, a beam of high-energy (hard) gamma rays is quite penetrating and may produce less biological effect in tissue than will a lower energy (soft) beam having the same total energy flux. The unit of tissue dose, the *roentgen*, denoted by r, is defined as that amount of X- or gamma radiation which will produce by ionization 1 electrostatic unit of electric charge in 1 cc of air under standard conditions. The details of this definition do not immediately concern us; the important point is that the roentgen is essentially a measure of energy absorption per unit volume.

By combining the definition of the roentgen with some experimental results, the fundamental definition can be converted into more useful forms. One roentgen is equivalent to

2.08×10^9 ion pairs/cc std air
1.6×10^{12} ion pairs/gm air
6.8×10^4 Mev/cc std air
5.2×10^7 Mev/gm air
83 ergs/gm air
91 ergs/gm tissue

The biological effects of high-energy radiations are much greater than would be anticipated from the amount of energy absorbed. The absorption of 1,000 r of radiation by the whole body would almost certainly be fatal, yet the temperature rise due to the absorbed energy would be only 0.002° C and only about 2 atoms out of 10^8 would be ionized!

It should be noted that the roentgen can be applied only to X- or gamma rays but particulate radiation can be treated by a logical extension of the original definition. A new unit, the *roentgen equivalent physical* (rep), is defined as that quantity of ionizing radiation which will produce 1.6×10^{12} ion pairs per gram of tissue or 83 ergs of heat energy per gram of tissue. As the name indicates, this is a purely physical definition and it does not follow that the biological effect of 1 rep will be equal to the biological effect of 1 r.

Biological effectiveness is introduced through a third unit, the *roentgen equivalent man* (rem), which is that quantity of radiation (of any type) which when absorbed by man produces an effect equivalent to the absorption of 1 r of X- or gamma radiation. This definition may be ambiguous since careful measurements may show that the relative effectiveness depends on the particular effect investigated. Any differences will probably be small and will not affect the practical application of the rem.

The roentgen is an unusual type of unit, representing as it does an energy absorption rather than a magnitude or an intensity. This has led to some confusion when the roentgen is applied to cases of human exposure. In this article the terms involved will be used in accordance with the following definitions:

Dose. The total quantity of radiation absorbed by the organism during a single radiation experience. Since the roentgen is a unit of energy absorption per unit volume or mass, we must specify the amount of tissue receiving the radiation before the dose can be determined. If one finger receives 1,000 r, the systemic effect will be much less severe than if the whole body

received 1,000 r. The same amount of ionization per gram of exposed tissue will be produced in the two cases, but the two doses will be quite different.

Dose rate. The dose received in a unit time.

Dosage. Strictly, this term refers to a regular schedule of repeated doses. It will be used here to denote the sum of the doses accumulated over a period of time. For example, a given operation may result in the operator receiving a whole-body dose of 0.01 r. If the operation is performed 15 times a day the daily dosage will be 0.15 whole-body r and in a five-day working week the dosage will be 0.75 whole-body r.

Exposure. The number of roentgens (or rem) with no reference to the amount of tissue receiving the radiation. From the definitions we have the relationship:

Dose = (Exposure)
× (Volume Exposed)

A discussion of methods of measuring radiation hazards is beyond the scope of this article but a few generalizations will be made. Biological injury depends upon the amount of tissue ionization produced by the radiation; hence, a good survey meter should give a response proportional to this ionization. Ionization chamber instruments can be designed to give correct readings of exposure or exposure rate over a wide range of energies and any quantitative surveys must, in general, be made with an ionization chamber instrument (*1*). Geiger-Müller counters, on the other hand, give readings which depend on the amount of incident radiation, with no reference to the ionizing power of the radiation. For example, if 100 beta particles of 0.15 Mev energy from C^{14} penetrate a G-M counter, the meter reading will be equal to that resulting from 100 3.5 Mev particles from K^{42} in spite of the fact that the latter have about 25 times the ionizing power of the former. Unless a G-M survey instrument is specially calibrated it should be considered as a sensitive radiation detector rather than as a measuring instrument.

External Hazards

In determining the safe limits for exposure to radiation, we must evaluate situations which have led to known injuries. Unfortunately, a good many cases of radiation injury have occurred, but equally unfortunate is the fact that in most of these cases there are no accurate data on the tissue doses. We must realize that any figures set for permissible exposures are still tentative, and are subject to modification as more information is obtained.

In the United States the permissible whole-body radiation dose has been set at 0.1 r per day for workers who may receive repeated doses in connection with steady employment. From the experiences of the Manhattan Project, it appears that 0.1 per day is a safe figure and one which need not be exceeded even when large quantities of active materials are being handled. Under unusual circumstances the permissible whole-body dose may be exceeded for one day if appropriate compensation is made on succeeding days, but under no circumstances should the whole-body dosage for any two-week period exceed 1.0 r. Future military operations in time of war may require whole-body doses considerably in excess of 0.1 r per day. Under such conditions excessive radiation exposures become part of the hazards of war.

Insufficient data exist for determining a lethal dose in man, but it is probably of the order of 450 r. Undoubtedly there are wide variations in individual sensitivities to radiation but little quantitative information is available.

There is some reciprocity between dose-rate and time. Thus, a whole-body dose of 0.1 r received over an eight-

hour working day appears to have the same effect as a whole-body dose of 0.1 r received, say, in 1 minute at the rate of 6 r per hour. It is not certain how far this reciprocity extends, and it is quite possible that a short flash of high intensity may produce more damage than the same dose delivered at a slower rate.

Internal Hazards

Radioactive materials may enter the body by ingestion, inhalation, or through cuts or abrasions. Once inside the body the fate of these substances depends upon the chemical properties of the particular element involved. Some elements, notably radium, plutonium and strontium, are "bone-seekers" and are stored in the bony structures; iodine concentrates in the thyroid and arsenic in the skin.

Radioactive materials introduced into the body decrease in activity with time because of radioactive decay at the rate characteristic of the element. Biological excretion reduces the amount present in the body at a rate depending upon the element and to some extent upon its location in the body. We can think of radioactive half-life and a biological half-life, the latter being that time required for the excretion of one-half of the original material. In calculating tolerances (*2*) the "effective half-life," which takes into account both the radioactive and biological half-lives, is frequently used.

Alpha particle emitters, which are of no concern as an external hazard, come into their own as an internal hazard. Radium, for example, deposits in the bony structures, and the emitted alpha particles are then effective in producing necrosis or osteosarcomas in the immediate vicinity. Gamma rays, which constitute the most usual external hazard, usually play a minor role as an internal hazard. Because they usually accompany beta or alpha emission and are not so strongly absorbed, they tend to produce only a fraction of the total ionization (*3*).

Permissible concentrations for internally stored materials are not established as satisfactorily as the standard for external radiation. The disastrous experience with the New Jersey dial painters indicates that 1 μgram of stored radium can produce serious injury (*4*), and 0.1 μgram is considered the tolerance value. K. Z. Morgan (*2*) has calculated permissible environmental concentrations from the requirement that no tissue receive an exposure of more than 0.1 rep of beta or gamma or 0.01 rep alpha radiation.

The determination of the amount of a stored radioactive material presents some difficulties. In general, these determinations must be made from the amount excreted in feces and urine and this usually requires a concentration of the desired element from large samples of excreta. Concentration is particularly required in determining alpha emitters for here the sample being measured must be very thin to avoid excessive self-absorption of the particles in the sample itself.

Biological Effects of Radiation

High-energy radiations are among the most insidious destructive agents for living tissue. Except for very large dosages the biological symptoms are delayed for periods ranging from a few days to several years. When an overexposure is detected through its biological effects there has already been serious tissue damage.

The most striking effects of overexposure are:

1. Leucopenia (decrease in the number of white blood cells)
2. Epilation (loss of hair)
3. Sterility (loss of ability to reproduce)
4. Mutations (altered heredity of offspring)
5. Cancer (abnormal cell growth)

6. Bone necrosis (destruction and death of the bones)

A decrease in the white cell count is one of the earliest signs of overexposure. A normal white count, however, is no guarantee that overexposure has been avoided, for substantial dosages are required and the effect may not appear for some time after the exposure. Conversely, a low white count does not necessarily indicate an overexposure since leucopenia can result from many other causes. However, if other factors are absent a series of low white counts in an individual exposed to radiation must be viewed with concern.

For many years X-rays have been known to produce epilation and have been used to remove superfluous hair. This has not always been accomplished without injury for the required dose is large. In some cases the hair may return after an interval of several months but with a sufficient dosage the epilation is permanent.

It is also well known that X-rays can produce temporary or permanent sterility. Cases have been reported where misguided individuals used X-rays as a means of contraception. In discussing radiation sterility with untrained workers care should be taken to distinguish between sterility, impotence, and libido, since only the first of these is directly affected by radiation.

Penetrating radiations are very effective in producing mutations, or changes in heredity. Mutants may not appear until the third or fourth generation, and a long time is required for the results to be known. The production of mutants is probably a chance phenomenon, with no threshold, so that any dose is effective with a probability that increases with dosage.

Radiation therapy is widely used in the treatment of cancer but we also know that cancer may follow excessive radiation exposure. Medical literature contains many case histories of radiologists who developed cancer before the dangers of X-rays and radium were recognized. While a moderate overexposure may result in a decreased white count, larger dosages may be followed by leukemia, a cancerous condition resulting in an enormous increase in the number of leucocytes. Post-radiation cancer may not develop until 10–15 years after the radiation, but there is no doubt of the causal relationship.

Bone necrosis is most commonly observed following the introduction of alpha emitters into the body. Although bone-seeking alpha emitters are the most effective agent for producing bone necrosis, cases have been reported following overexposure to other radiations.

Many other biological effects of radiation are known but cannot be discussed here. More complete presentations are to be found in the literature (*5*, *6*). An interesting effect of possible diagnostic value is the production of abnormalities (7) in the finger ridges.

Waste Disposal

The problem of the disposal of radioactive wastes is one of the most important that confronts the health physicist. Once radioactive solutions have been dumped down the drain they are gone beyond recall and it cannot be assumed that they will always accommodatingly go to inaccessible places or will be sufficiently diluted to be harmless.

Burying might seem to be a safe method of disposal but this may be dangerous under some circumstances. The active material may come in contact with ground water and be so distributed as to pollute streams and sources of drinking water. Incineration also is not satisfactory, particularly with long-lived isotopes. Many solutions to the problem of waste disposal have been suggested but none seems to be ideal. It may be desirable to institute a

nationwide collection scheme with the ultimate disposal burial at a great depth in the sea or in a national burial ground.

In view of the unpredictable but undoubtedly large number of future uses of radioactive materials, it is imperative that every effort be made to avoid widespread contamination, even of a low order. Too little is now known about the biological effects in man to risk a large scale pollution which would be difficult or impossible to eliminate.

Care must be taken in the disposal of even small quantities of active materials. At the end of the war there was a large surplus of instrument dials painted with radium-containing luminous compounds. If these had been turned in to the scrap metal industry, the dilution would probably have been great enough to preclude radiation injuries, but any instruments made of this metal for detecting radioactivity would have shown a serious increase in background counts. A similar situation might easily arise in the scrap paper industry with serious consequences for the manufacturers of photographic materials.

Role of the Health Physicist

The health physicist is a vital link in any operations involving the use of radioactive isotopes or sources of high-energy radiations. Unskilled workers, ignorant of the dangers involved, must obviously be protected from noxious agents which they cannot detect with any of their senses. A more difficult job is the protection of skilled workers, who understand the hazards, but who have an interest in the use of the radiations. Every health physicist is all too familiar with the enthusiast who omits precautions and gets a substantial radiation dose for which he hopes to compensate by avoiding all exposure for some time thereafter. This is a dangerous practice, for this type of individual is usually the one who subsequently forgets to take the promised radiation "vacation."

One of the major responsibilities of the health physicist is the education of the personnel under his protection. Here he must adopt a middle course, emphasizing that radiation is potentially dangerous but quite safe if properly handled. He must point out that the establishment of a permissible exposure level of 0.1 r per day does not grant a license to receive this amount with impunity. It is important to stress that any amount of radiation is undesirable and exposure should be avoided as much as possible.

The health physicist must be equipped with the proper types of survey and laboratory instruments, together with means for repairing and calibrating them. The excuse that a small laboratory does not have a sufficient budget for such items is not a valid one. Radiation is no respecter of budgets and will injure the rich and the poor alike. Responsibility for keeping exposure records also rests with the health physicist. As more persons come in contact with radiation it becomes increasingly important to know past exposure records. Obviously these records must be available for long periods of time because of the delayed effects of overexposure.

The health physicist should have complete authority over all procedures involving radiation hazards. Such authority does not imply a restriction as to what may or may not be done, but it does prescribe control over detailed methods and procedures. This control need not hamper investigation, but can actually aid it, since responsibility for health protection is transferred from the investigator to the health physicist.

There are three general methods by which external radiation hazards can be reduced: increased distance from the source, increased shielding, and decreased time of exposure. A com-

promise is frequently involved since increased shielding or distance may result in a disproportionate increase in time. The use of mechanical manipulators for working at a distance should be encouraged. The construction of suitable gadgets frequently calls for the expenditure of considerable ingenuity and time but should be insisted upon whenever necessary. Equally important is adequate practice with non-active materials, so that smooth operation is assured when the active material is present. As an example of what may be involved we cite the case of a chemical reaction which required 15 minutes to carry out. After careful study, more than one month was spent in making remote control devices and practicing with them. Then, using active materials, the reaction was carried out safely in 15 minutes.

The health physicist can carry out his work most successfully if he has no direct connection with the experiments. If he is directly involved his enthusiasm to get results may lead him to take unnecessary chances for the sake of completing an experiment. By placing health protection in the hands of an outsider who can make an objective, unemotional analysis of the situation, considerable overexposure can be avoided.

Obviously the responsibility of the health physicist is great. It rests with him as to whether or not we use these most important and potent radiations without untoward effects in this or in later generations. With proper training and with authority to enforce the necessary safety rules, he will be able to carry out his mission successfully.

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The Fission Products and Their Uses

A brief, historical review of the developments leading to the present-day knowledge of fission and fission products. Applications of bulk and separated fission products are outlined

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IN 1934, NOT LONG after the discovery of the neutron by Chadwick, Fermi (*1*) and his collaborators, working at the University of Rome, bombarded uranium with neutrons in an attempt to produce transuranic elements, *i.e.*, elements with atomic number Z greater than 92. By analogy with previous work they expected the compound nucleus formed by neutron capture to be unstable and to revert to stability by the emission of a beta ray, thus increasing the atomic number by 1 unit.

They obtained activities which, by virtue of half-lives and energy, did not correspond to the then known so-called "natural" radioactive series. As indicated by reference to the periodic table, it was expected that at least one of these activities would be homologous chemically with either Mn, 43, or Re. The activities were co-precipitated with MnO_2, and, on the chemical evidence thus obtained, it was concluded that elements with $Z > 92$ were found.

It was pointed out by Noddack (*2*) that MnO_2 had very great scavenging properties, and she criticized the conclusions which were drawn on such scanty chemical evidence. She pointed out that the uranium nuclei might have split and the scavenging effect of the MnO_2 was bringing down elements in the middle of the periodic table. Unfortunately no supporting evidence was presented with this argument and it was not seriously considered, although the criticism of the chemistry was certainly valid.

This work was continued by Hahn, Strassmann and Meitner (*3*) at the Kaiser Wilhelm Institute in Berlin using uranium and thorium. Soon an entire host of so-called transuranic elements made their appearance, some of which were difficult to reconcile with the expected properties of the transuranics. Several radioactive chains were found. Some of these started with elements which would co-precipitate with Ba and were thus ascribed to Ra. The daughter elements had the chemical characteristics of La and were thus characterized as Ac. The Ra was said to have been formed from ${}_{90}Th^{232}$ by an (n,α) process or from the emission of two successive alpha particles from the excited ${}_{92}U^{239}$ nucleus formed by neutron capture. However, a multiplicity of so-called "radiums" made their appearance and they became more and more difficult to reconcile from a nuclear point of view. It became very difficult to explain, for example, why the ${}_{92}U^{238}$ nucleus which is ordinarily stable should on neutron capture become excited violently enough to emit two successive α particles.

Curie and Savitch (*4, 5*) focussed their attention on an activity with a 3.5 hour half-life which had been characterized by Hahn as actinium. Fractional

precipitation of the oxalates showed the element to be more nearly identical to La in its properties than to Ac. If this were really La, its precursor would be not Ra but Ba. Hahn (*6*) carried through very stringent tests to prove or disprove this and came to the conclusion, by reason of fractional precipitation with Ba and Ra, that the activities which he had previously described as Ra were in reality Ba. Re-examination of all the "transuranics" showed that they could all similarly be placed in the middle of the periodic table. Eka-Pt, for example, was found to be iodine. As a carrier, to precipitate the platinum homologue, Hahn had used a well-known reagent, tetrammine platinum chloride $Pt(NH_3)_4Cl_2$. The carrier really brought down the active halogen, not the transuranic element.

These reactions are now understood to occur as follows:

$$_{92}U^{238} + {}_0n^1 \rightarrow {}_{92}U^{239} \xrightarrow[23.5\ m]{\beta^-}$$

$$_{93}Np^{239} \xrightarrow[2.3\ d]{\beta^-} {}_{94}Pu^{239} \xrightarrow[2.44 \times 10^4\ y]{\alpha} \quad (1)$$

$$_{92}U^{235} + {}_0n^1 \rightarrow {}_{92}U^{236} \rightarrow$$

$$z_1A^{m_1} + z_2B^{m_2} + V({}_0n^1) + K.E. \quad (2)$$

In the first reaction the uranium undergoes radiative capture to form a uranium isotope, unknown in nature, of only transitory existence, which by β^- emission then proceeds to form genuine transuranic elements with $Z = 93$ and 94.

The U^{235} nucleus, present to 1 part in 139 in natural uranium, is unstable by about 160 Mev towards fission. However, a potential barrier of 5–8 Mev must be crossed before such fission will take place. This energy fillip is provided by the captured neutron. An excited compound nucleus $_{92}U^{236}$ is formed which has a very short lifetime, splitting into two fragments of roughly equal size.

Detailed investigations (*7*, *8*) have yielded the following information regarding the fission process: In equation 2, V, the number of neutrons per fission, is between 1 and 3. The kinetic energy liberated per fission is of the order of 160 Mev. The values of Z_1 and Z_2 have been found to be between 30 and 62. Thus, in fission, every element between Zn and Eu is found. These fission products are formed with a great deal of excess energy and some of this energy is dissipated in the ejection of the fission neutrons and gamma rays. Most of the energy is used up in the kinetic energy of the fragments which have very high velocities, approaching $\frac{1}{10}$ that of the speed of light. Due to the great neutron-over-proton excess (or mass-to-charge ratio), the products also contain considerable nuclear potential energy which disappears in successive β^- decays with the formation of radioactive chains. A total of approximately 160 active isotopes, distributed among 64 chains, have been found. There are, on the average, about 3 β^- decays per chain before stability is reached. Several investigators (*9*, *10*) also reported α tracks found during fission. No case of ternary fission has yet been substantiated.

From energy considerations one would expect fission to occur symmetrically, or at least for that mode to predominate, since the activation energy for symmetrical fission is less than that for asymmetric. From a consideration of the fission yield, however, it can be seen that this is not the case. The fission yield of an isotope is defined as the probability of that isotope's being formed in fission. This can be determined in several ways. Two samples of U^{235} are irradiated in the pile. One of these is very thin and is mounted in a fission chamber (*11*) and the total number of fissions occurring in the sample determined. The other, a thicker sample, is dissolved after irradiation and a chemical separation

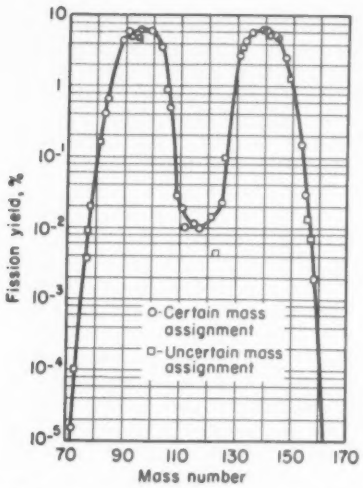

FIG. 1. Yields of U^{235} fission product chains as a function of mass

of one element performed. Ba^{140} is usually selected for this purpose because of its high yield, convenient half-life and simple chemical separation. The percentage of fission leading to that nuclide is then determined. This can then be used as a reference, and, with the total fission determined from the other sample, the fission yields of the other nuclides found.

The direct measurement of the number of U^{235} thermal neutron fissions can be avoided if natural uranium is bombarded with thermal neutrons (*12*). In this case, as shown in equations 1 and 2, U^{238} undergoes radiative capture to form U^{239} whereas the U^{235} undergoes thermal neutron fission. The β^- activities produced both by capture and by fission in natural uranium during thermal neutron bombardment are proportional to the respective cross sections for these processes. Thus, at saturation of bombardment,

$$\frac{\beta^- \text{ activity of } U^{239}}{\beta^- \text{ activity of fission isotope}} = \frac{\sigma_c}{Y_f \sigma_f}$$

where σ_c and σ_f are the capture and fission cross sections, respectively, for thermal neutrons in natural uranium and Y_f is the fission yield of the isotope studied. Once the fission yield of a nuclide, like Ba^{140}, is determined, then it is used as a reference as above.

In Fig. 1 is shown the fission yields of the various chains as a function of their mass number (*7*). Asymmetric fission occurs with the formation of two groups—a light group and a heavy group. Recent work by Perlman, *et al.* (*13*) and Turkevich (*14*) has shown that the tendency towards asymmetry disappears as the neutron energy increases. The light group has been shown, by chemical means, to contain all elements lying between Zn and Pd while the heavy group contains all elements between Pd and Gd. There is evidence for all mass numbers from 72 to 158. Ninety-seven percent of the fissions give rise to mass values in the light group of 85 to 104, with atomic numbers 30 to 46, associated with values in the heavy group of 130 to 149, with atomic numbers 46 to 72. The most probable ratio of fission product masses is 95/139.

The half-lives of the nuclides produced in fission vary from very short, of the order of fractions of seconds, to very long, of the order of hundreds of years. The energies associated with these radioactive elements vary from very soft β's and γ's to β's with maximum energy of 5.3 Mev and γ's of about 2 Mev.

Up to 1942 the major sources of neutrons were the cyclotrons or Ra-Be, and the fission products produced presented no disposal problem. However, in the production of plutonium, or in the use of atomic power, fission products are a necessary byproduct. Methods have been evolved for the extraction of the uranium and plutonium leaving the fission products in a large volume of solution along with residues from the extraction processes. The disposal

problems arising from these highly active solutions is a great one. The fission products, as has been shown, contain many long-lived components and it would take many years before the solutions could be safely diverted into normal disposal avenues. The solutions could be stored in specially shielded tanks, but this presents a major space problem. Emptying solutions into rivers after a decay time long enough to conform with local health regulations could possibly be done. This, however, is highly wasteful of materials which may be useful.

Fission Product Uses

A brief outline of projected uses for the fission products follows. This is not intended to be original. Many of the suggestions have come from coworkers during numerous discussions and the origin is very difficult to trace.

The production of plutonium for military and industrial purposes has necessitated the expending of a very great effort to gain general and specific knowledge of the chemistry of the fission products. Some are easily separable from the general group by the classical methods of inorganic chemistry, complicated of course by the necessity of remote control operation. Others can be separated by radioactive decay methods—separation of a parent nuclide which by decay then forms a daughter which is not too easily separated. A very powerful method for the separation of fission products is by adsorption on ion-exchange resins (*15*). The fission products are adsorbed on a very thin layer in the ion-exchange column and, by elution with various complexing agents at various pH's, a very good separation is obtained. On the assumption that it is possible to separate from the fission products all contaminating materials used in the plutonium and uranium separation processes, the remaining solution can be concentrated, leaving the fission products in a very small bulk. Thus, in considering the uses, we should consider both bulk fission products and separated ones.

For certain types of medical therapy, very compact telecurie sources containing many curies per cc are needed (*16*). Also sources for interstitial use require linear densities of the order of 1 mc per cm of source. Particularly for nonremovable sources, very high concentrated materials of the order of 100 curies per cc are desirable. In such cases gaseous products have hitherto been used. However, experience has shown such sources to be undesirable, except for special containers of complex shape which would be difficult to fill uniformly with nongaseous radioactive material.

Because of the great variations in β^- and γ energy and variations in half-lives, the fission products can very conveniently serve as therapeutic agents. Theoretically, several hundred curies can be concentrated in one gram of solid. An obvious difficulty is the transport and handling of such large sources as a matter of routine, but the amount dispatched to any therapeutic center might only be that required to raise the strength of source in use to its original value. The source of radiation can be U metal blocks irradiated in a chain-reacting pile for a suitable period and then allowed to "cool" for a few months. Under these conditions it should be possible to prepare a compact telecurie source of, for example, 100 curies. Since this source would have a half-life of only a few months, depending on the length of time of irradiation and cooling, it would have to be reactivated or replaced periodically. After an extended period of time, these blocks could be returned for Pu extraction. The self-absorption in a metallic uranium source would be serious unless the U were irradiated and

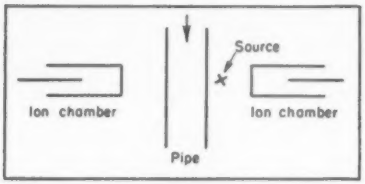

FIG. 2. Determination of fluctuations in density of a liquid moving along a pipe

used in a form having large surface area per unit mass. The advantage of such a source is the relatively enormous quantity of radioactive material available easily and cheaply and the absence of costly and dangerous chemical separation. Much experimental work on the therapeutic side has to be done on the nature and penetration of the mixed radiations.

Since the fission products contain such a diversity of radioactive substances emitting a large variety of β^- and γ energies and possessing such widely different half-lives, it should be possible to pick out a particular nuclide with specific therapeutic or diagnostic use. In some cases, high-energy beta-ray emitters free from gamma rays are necessary so as to ensure reasonable penetration of surface lesions, with a sharp decrease of dose at a depth equal to the range. In other cases gamma rays are convenient as they add to the local beta absorption.

It has been shown by Mayneord and others that a one millicurie point source of gamma rays of energy 0.4 Mev gives 2.0 r per hr at 1 cm distance; 100 curies, *e.g.*, of I^{131}, gives a dosage rate of 2.0×10^5 r per hr at 1 cm. Even at 5 cm we would expect some 130 r per min. This implies that such a source might be used conveniently in place of low-voltage X-ray equipment in the treatment of cavities (*e.g.*, mouth, vagina, rectum). Because of the low energy of the gamma rays from I^{131} with lead shielding, it should be possible to design sources of small linear dimensions with simple shutters to control exposure and thus to obtain a very simple, compact and useful addition to therapeutic equipment. Other fission products might lend themselves to similar techniques.

In addition to the above, certain elements are specifically absorbed by various parts of the body. The uses of Sr^{89} in biological experiments because of its selective deposition in bone and I^{131} in investigation and treatment of the thyroid are too well known to require further explanation. It may be that investigation will show that other such nuclides found in fission may be useful.

In addition to the uses in the medical field, fission products may be used industrially. For example bulk fission products may be used in control procedures. A change in the density of a solution moving along a pipe might be detected as shown in Fig. 2.

Two ionization chambers are connected differentially on opposite sides of a pipe through which a liquid, in which the density change is to be determined, is flowing. A source of bulk fission products is used, the decay being automatically corrected for by the differential ion-chamber set-up.

Similarly the interface between two liquids in a column might be detected by remote control by the use of a Geiger counter as a probe. A radioactive indicator is chosen which is soluble in only one of the liquids and, when the liquid passes the monitor, an alarm can indicate the fact. Similarly the probe may be movable.

Bulk fission-product sources can also be used to replace Ra and low voltage X-ray sets for use in radiography. Some of this work has already been done by Tenney (*17*) using La^{140}, the 40-hour half-life daughter of Ba^{140} (12.8 day half-life). He points out that it is conceivable that some day even the

short-lived radioisotopes may be used for this purpose, since on the basis of simple economy, where only a few radiographs are needed, these may prove themselves to be cheaper than Ra or X-ray equipment.

Many miscellaneous uses suggest themselves. Beta emitters can be used in the preparation of luminous paints. They can also be used on spark plugs or gas discharge lamps to reduce the striking voltage. One of the hazards in grain elevators, for example, is a dust explosion caused by electrostatic charges picked up by moving belts. Fission products would discharge them. A very minute source placed in a balance case would have the same effect there. Bulk sources could also be used to help trace the movements of underground waters, air masses and oil beds. A suggestion has also been made that valuable objects placed on exhibition, such as paintings, for example, could be easily marked with fission products so that the unauthorized removal of same would set off an alarm. Obviously many more similar uses of bulk fission products can be suggested.

There are also many uses for individual fission products. Marinsky, Glendenin and Coryell (*18*) show that considerable amounts of elements 43 and 61, the existence of which was hitherto doubtful if not nonexistent, can be prepared in the chain-reacting pile. A uranium pile operating at the power level of 1000 kev produces 3×10^{16} fissions per second. Since 2.6% of these, for example, lead to 61^{147}, 16 mg of 3.7-year element 61 is formed after suitable decay. Similar amounts of element 43 will be formed, and the physical and chemical properties of these two elements can be determined.

Some nuclides are produced in fission which are not easily produced by other neutron reactions, *e.g.*, Ru^{106}. The study and/or use of these necessitates their separation from fission products.

Pure stable isotopes can be prepared by separation of some of the individual members of the fission chains and allowing them to decay to the stable end-member.

Some of the fission products have γ's of sufficient energy so that combined with Be they would make good photo-neutron sources, *e.g.* La^{140}.

It would be impossible and superfluous to list all of the uses of the separated fission products as tracer elements in research. Enough has previously been written about this to make the applications obvious. However, the above is a very general attempt to broadly outline but a few of the many uses for both bulk and separated fission products.

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To get around this language barrier, Brookhaven National Laboratory has begun to compile, in the form of title lists, a "Guide to Russian Scientific and Technical Periodical Literature." In an Introductory Note to the "Guide," H. H. Goldsmith, head of Brookhaven's Information and Publication Division, acknowledges the translation work of Dr. Ludmila Turkevich of the Modern Language Department, Princeton University, the advice of Professor John Turkevich of the Chemistry Department, Princeton University, and the active cooperation of Dr. S. C. Stanford, Technical Librarian of Brookhaven National Laboratory, without which this important project could not have been started.

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* Published in 1948

The Future of Nuclear Power

AFTER THE DRAMATIC and explosive climax to the war at Hiroshima and Nagasaki, which overshadowed in their political and social implications all the previous events of the war, the layman enthusiastically adopted an optimistic appraisal of the immediate promise of the Atomic Age. The pill which dropped in a gas tank would run an automobile forever, the drop of water which would drive a transatlantic liner, the sensational cancer cures, the rockets to the moon, free electricity—all these oft-repeated wonders made a deep impression on the public mind. It was only after considerable effort that responsible scientists and engineers succeeded in establishing a more reasonable view of the potentialities and dangers.

One can, therefore, easily understand the extreme caution which characterizes official pronouncements in all countries concerning the probable future of nuclear power. This understandable caution, unfortunately, carries the possibility of inducing a defeatist attitude about the ultimate benefits to mankind of the new discoveries. A more subtle danger is the diversion of effort from long-range projects, which are not too well appreciated by an impatient public, to concentrate on easily attainable immediate results.

The pendulum of public opinion has now swung so far the other way that one hears commonly expressed the view that there is no real need for nuclear power in the United States. This narrowly provincial view, which ignores the power famine in the rest of the world—a power famine so great that its resulting limitation on productivity starves to death far more people than any medical advance through radioisotopes can hope to save—is not even correct for our own country.

A recent article by R. C. Gunness of the Standard Oil Company of Indiana in *Chemical and Engineering News*,* points out the remarkable fact that the entire increase in fuel consumption in the United States during the last 25 years has been due to increased use of petroleum. The extraction of coal during this period has been static—coal production has equalled the level of 1920 only in two years, 1944 and 1947. At present the private utilities industry is in the middle of the largest construction program in its history, comprising three million installed kilowatts per year. From the standpoint of conservation of natural resources, it is unwise to extend further the replacement of coal by heavy fuel oil, which is a marginal and uneconomic use of petroleum. Where, then, will the increased supplies of fuel come from to support our expanding industrial frontiers and increased use of electricity on the

* *Chem. Eng. News* **26**, 2123 (1948).

farm and in the home? Who can be so optimistic to think that we will always have convenient rivers to harness if even the West Coast is experiencing a power shortage?

The most important question about the use of nuclear power is its economic feasibility. The only published economic evaluation is that released by Mr. Baruch to the U.N. Atomic Energy Commission. This showed that nuclear power required a high initial investment, but still could compete in the United States with coal priced at more than ten dollars a ton. This certainly seems an attractive proposition for European nations where coal is in excess of fifteen dollars a ton. Further, it must be remembered that the Baruch figures were based on the Hanford-type pile, which was admittedly a wartime improvisation. Whether any more up-to-date estimates are available is not clear. The AEC General Advisory Committee has reported two economic forecasts, one based on favorable, and the other on unfavorable assumptions, about the cost of uranium and the technical practicability of breeding. It goes on to say, "At the present time, sufficient knowledge does not exist to make a definite choice between these two alternative possibilities."

It is hard to see how so vague a statement can give the American public or the world any measure of the true potentialities of nuclear energy, or any adequate basis for determining intelligent domestic and foreign policies. Certainly one would have hoped that at so late a date—it is now two years since the Baruch Nuclear Power Report was released to the U.N. AEC on September 5, 1946—the U. S. AEC would have made better progress in determining, in the words of the McMahon Act, "the social, political, economic, and international effects of" . . . "industrial, commercial, or other non-military use of fissionable material or atomic energy."

The next most important consideration is how soon we may expect to reap any positive return in the form of useful energy from our huge investment. The Fourth Semi-annual Report of the U. S. AEC expresses the opinion that it would be twenty years before "any considerable portion of the present power supply of the world" will be generated by nuclear energy. Of course, the time scale for engineering progress is one of the most difficult items to gauge, depending as it does on so many imponderables, such as the enthusiasm, determination, talent, and manner in which it is attacked. But certainly no one can disagree that, at the present rate of progress, it will be at least twenty years before appreciable results are obtained.

—*Walter M. DeCew*

NUCLEONIC EVENTS

ANALYZE FUTURE STATUS OF NUCLEAR POWER

Ed. Note: Below are two analyses of the future possibilities of nuclear power. The first is by Robert Colborn of the McGraw-Hill Washington Bureau and is based on the Fourth Semiannual Report of the U. S. Atomic Energy Commission. The second is based on an interview with Sir John Cockcroft by the McGraw-Hill World News Bureau. For editorial comments on the future of nuclear power, the reader is referred to page 76.)

Ultimate output of atomic energy in this country may be as great or greater than present output of coal power "and will operate at a lower cost, at least as far as fuel expenditure is concerned."

That's the Atomic Energy Commission's latest view on power prospects, provided that favorable assumptions are made about two things:

1. That the cost of uranium does not rise markedly in the future because of forced resort to lower-grade ores; and

2. That the theoretical possibilities of "breeding" atomic fuel can be realized in engineering practice. This last means building atomic reactors which are highly efficient—so that when they consume fissionable materials they will at the same time produce an equal or greater amount of new fissionable material by transmutation of non-fissionable uranium or of thorium.

If unfavorable assumptions are made —that the cost of uranium will rise and that breeding will prove impracticable —then atomic power could compete with coal power only in regions where cost of transportation from the mine is a determining factor.

This view on economic prospects from atomic power is much the most optimistic which has ever come from official sources. It puts the official AEC position much more nearly in line with private estimates that the ultimate cost of atom power would drop somewhere between hydro and steam.

The new position appears in the fourth semiannual report of the AEC. It is presented as a report to AEC of the commission's General Advisory Committee. This source is particularly significant, since the advisory committee has been generally considered as the group in the government most pessimistic on power possibilities.

As to time scale, the advisory committee is still rather conservative. It points out that two reactors to produce token electric power should be completed within two or three years.

One of these is the air-cooled natural-uranium pile at Brookhaven Laboratory on Long Island, due for completion early next year; it will produce some power from the heat of its cooling air but not even enough to run the blowers which drive the cooling system.

The other is a reactor of advanced design at the Argonne Laboratory in Chicago. This will be a fast-neutron pile using enriched fuel and cooled by liquid metal. It will run at a high temperature and will produce power in something approaching commercial quantities, though hardly at commercial costs. Construction of this reactor, under discussion for the last year and a half, was finally given formal approval in June.

Building on experience with these two units and their successors, the advisory committee thinks, "fairly practical

reactors that might be useful for special purposes" should be available within a decade.

In about 20 years, under favorable circumstances, a "considerable portion of the present power supply of the world" might be replaced by nuclear fuel, the committee thinks.

* * *

Nuclear energy will not contribute any very large amount of power to the world during the next ten years. That is the opinion of all well-informed scientists now working in this field, according to Sir John Cockcroft, director of Britain's Atomic Energy Research Establishment.

Reason: four major technical problems still have to be solved.

1. The "fuel" problem—investigations so far have shown that, while natural uranium piles which could generate power as a byproduct are technically feasible, their "fuel" efficiency would be so low that they would not be economical. Natural uranium piles "burn up" only a very small proportion of the uranium. The fuel problem thus becomes the so-called "breeding" problem: finding a way to utilize the greater part of the uranium "fuel" consumed in the pile. Primary fuel source, uranium-235, must be made to breed secondary fuel from U^{238} or thorium. Until this is done, present resources of uranium will be insufficient for large-scale nuclear power.

2. The metallurgical problem—finding materials, better suited to withstand high temperatures, which can be used in construction of piles.

3. The "combustion" problem—finding an efficient way to process the nuclear fuel used in the pile, which includes removing the intensely radioactive ash from the fuel safely and economically.

4. The "ash disposal" problem—getting rid of the large quantities of the radioactive ash that would be produced by the sizable number of nuclear power stations that would be required to supply the world.

BRITISH ATOMIC SCIENTISTS ISSUE STATEMENT ON FAILURE OF BARUCH INTERNATIONAL CONTROL PLAN

The following statement was issued by the Council of the British Atomic Scientists Association, July, 1948.

1. The Atomic Scientists' Association since its foundation in 1946 has pressed for international control of atomic energy. It has become obvious in recent months that this is unlikely to be achieved in the near future and in view of the recent proposals by the British, French and U. S. Governments to adjourn the Atomic Energy Commission of the United Nations Organization, it is clear that the attempt to achieve it has for the time being failed. A restatement of the position of the Association in this matter is therefore necessary.

2. In 1946, when the work of the Association began, it seemed that the existence of machinery for the control of atomic energy could do much to lessen suspicion and decrease the dangers of war, and also that the recent invention of atomic weapons could be used to induce the powers to set up such a scheme. The reasons for this belief were the following:

(a) The destruction in the cities of all belligerents which the strategic use of atomic bombs would entail, and the difficulty of envisaging a satisfactory system of defense, provided obvious incentives to the setting up of a

control scheme.

(*b*) The technical possibility of the control of atomic energy, as described in the Lilienthal report, was widely held to make it a suitable subject for a first exercise in the control of armaments.

3. It is now clearer than it was in 1946 that the sources of disagreement between the major powers are so deep that the institution and working of a control scheme would be much more difficult than at first supposed. As a result there does not appear to be any chance of agreement at the moment either on the lines of the Lilienthal proposals or on any more limited scheme. The reasons for this disagreement can perhaps be summed up as follows: The United States Government would be unwilling to abandon its lead in atomic weapons without a complete system of inspection and control which would ensure that no other country was building up these armaments to use against America. The U.S.S.R., on the other hand, believes that in any control authority acceptable to the Americans the countries voting for Russia would be a minority, and that the development of atomic energy in their country would pass under foreign direction, with resultant foreign influence on their whole economy. In addition, their attitude may be influenced by the view that any complete scheme of inspection would give the outside world information of strategic value about the location of their industry.

4. The Council of the Association wishes to put on record its continuing belief in some form of international control of atomic energy as the most desirable ultimate solution of the problem of atomic weapons; it does not, however, consider that any good purpose will be served by pressing for it now. A substantial change in international relations is necessary before such plans—in a form which will have to depend on the international situation at the time—can be revived with any hope of success; it is possible they will then be linked with proposals for limitation of other weapons, and, in a changed situation, some benefit might perhaps be expected even from a scheme less complete than the Lilienthal proposals.

5. Certain other solutions have been put forward from time to time. One of these is that a moratorium should be established in the development of atomic energy, so that the nations of the world would agree to build no large scale plants capable of producing fissile materials. This would not only stop work on essential power production, but would be meaningless unless it included the closing-down of American production plants and disposal of stocks of fissile material. This would not be acceptable to the Americans without the same safeguards as have been proposed in the control scheme. Such a scheme would still involve the problem of inspection to ascertain the absence of undeclared installations, and this, rather than the inspection of declared plants, seems to lead to most serious disagreement. For those reasons, the Council does not believe that a moratorium offers any hope of easing the problem.

6. Another suggestion that has been made is that a control scheme should be set up, which did not include all nations, but to which all nations willing to do so could belong. We believe that this proposal misses the point of the original plans for international control. These were designed to promote agreement and remove suspicion between nations which might otherwise be hostile. One important feature of the control schemes was that once a system of safeguards had been set up, there would no longer be any need for secrecy in scientific and technical

matters, and this would make an important contribution towards removing mutual suspicion. It is certain that in a scheme which did not include at least all major powers secrecy would be retained. Schemes for closer collaboration between some nations, for example, between Western Europe and the United States, might, of course, be desirable because of their advantages in the economic and military spheres, but they should not be regarded as a step towards the international control of atomic weapons.

7. The Council, seeing no hope of an early agreement regarding the atomic bomb and other weapons of mass destruction, believes that the solution can now be sought only through the constant promotion of all measures designed to make war less likely, and to reach a settlement between East and West. In particular, it does not believe that any solution tolerable to the peoples of this country and of Western Europe could be obtained through the dominance of one side over the other. It does not believe that through the development of atomic weapons alone any nation is likely to be able to secure quick victory over a great power with widely dispersed industries; any conflict would be a long and costly struggle, in which Western Europe is likely to be a battlefield and in that case would suffer serious devastation.

8. The Council of the Atomic Scientists' Association also wishes to reaffirm its belief that collaboration between scientists of different nations is of value both in the interests of science itself and as a contribution towards international understanding. In particular, in view of recent political developments, we believe it important to maintain and improve contacts between the scientists of countries in Eastern Europe and those of Western Europe and America.

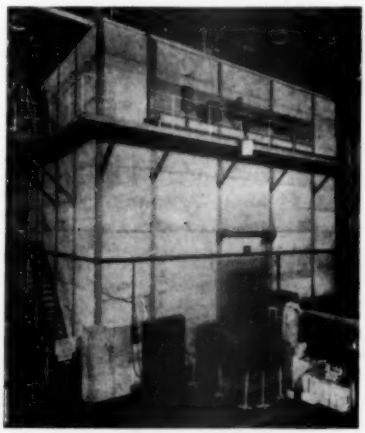
BRITISH OPERATE TWO PILES AT HARWELL

The British now have two piles in operation. The GLEEP (graphite, low-energy, experimental pile), which develops about 100 kilowatts of power, went into operation in August, 1947. In July it was joined by BEPO (British experimental pile), which will develop about 6,000 kilowatts.

These facts were included in the review of research progress which was outlined before members of the press in their first visit to the Atomic Energy Research Establishment at Harwell, England, in July.

Also included in the progress report were the following:

Fundamental research on nuclear physics will go ahead faster as soon as several different types of accelerators for nuclear particles now under construction are completed. One of these, a 5-million-volt Van de Graaff generator



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General view of GLEEP. The holes through which samples are put into the pile for production of radioisotopes are in the face on the right. In the same face a square screen of lead blocks covers the thermal column. The latter is a column of graphite used to produce slow, or thermal, neutrons for experimental work

used to accelerate hydrogen nuclei, is already complete at Harwell. Another, a 110-inch cyclotron, with a magnet that weighs 700 tons, which is capable of accelerating protons or deuterons to 200 Mev, is expected to be running by the end of this year. Linear accelerators are being built by a Harwell unit working at the Telecommunication Research Establishment at Malvern. There, also, was recently completed a 30-million volt synchrotron, which is the prototype for a 300-million volt unit subsequently to be constructed.

By last spring, the GLEEP had produced 150 specimens of radioisotopes, production of which was begun in September of last year.

Separation of stable isotopes is being undertaken by several different processes, including thermal diffusion, electromagnetic separation, chemical exchange, and fractional distillation. An experimental plant for the production of carbon-13 by the last-named method has been built, and, by the end of the year, a plant should be in operation which is expected to be capable of turning out C^{13} at the rate of 0.3 grams per day, sufficient to meet present British research demands for this substance.

MIT GETS GRANT TO STUDY TRACERS IN METALLURGY

Research on the application of radioactive tracer techniques to mineral engineering problems will be expanded in the Department of Metallurgy at the Massachusetts Institute of Technology under a grant from the research division of the Atomic Energy Commission, according to a recent announcement by Thomas K. Sherwood, dean of engineering.

The project, which will be started at once, will be under the direction of an advisory board composed of B. Old of Arthur D. Little, Inc., and the AEC; A. M. Gaudin and R. Schumann, Jr.; J. Dasher of the Division of Industrial Cooperation; R. D. Evans of the department of physics; J. W. Irvine, Jr., of the department of chemistry; and M. J. Buerger of the department of geology at MIT. Professor Gaudin is supervisor of the project, and H. Rush Spedden is its executive director.

BROOKHAVEN BUILDS WEATHER TOWERS FOR PILE

Two meteorological towers for weather observations in connection with nuclear research are being built at Brookhaven National Laboratory, Upton, N. Y., it was recently announced. The towers (one, 420 feet high, and the other, 160 feet high) will be used to furnish information on wind currents affecting the operation of the nuclear reactor which is expected to be finished this fall.

The Brookhaven reactor will be cooled by air. Large fans will conduct the air away from the pile, through an air duct, and up a 300-foot air stack on a hill near the reactor. In the air will be minute quantities of radioactive argon which is expected to be dissipated harmlessly into the upper air. Study of wind velocity and direction, and atmospheric pressure, during operation will permit control of the pile so that air emanating from the stack will not settle on or near the ground.

$250,000 GRANT TO MIT FOR FISSION RESEARCH

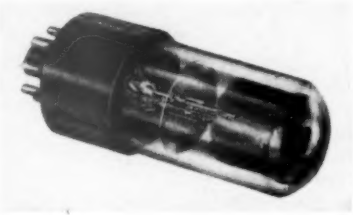
A grant of $250,000 for basic studies of nuclear fission and its engineering has been made by the Socony-Vacuum Oil Company, Inc., to the Massachusetts Institute of Technology. This work will be carried on principally in the MIT laboratory for nuclear science and engineering. The grant will be made at a rate of $50,000 per year for five years. It will be utilized at the school's discretion to provide facilities and scientists for carrying out the actual work involved in solving fission problems.

PRODUCTS and MATERIALS

THORIUM DETECTOR

A Geiger counter for the determination of the thorium content in thoriated tungsten wires has been developed by **Amalgamated Wireless (Australasia), Ltd.**, but it has been emphasized that the instrument is not in commercial production.

Wires containing a percentage of thoria or thorium oxide are used in radio transmitting tubes to obtain a



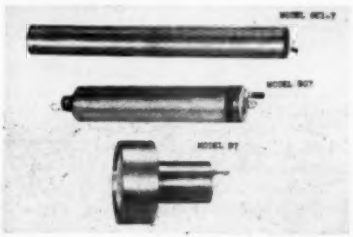
useful electron emission at lower temperatures than would be possible with straight-tungsten wires. To meet the need for quickly identifying the various types of wire used in the construction of such tubes, the special Geiger counter has been devised in conjunction with a recording unit. This consists of a radio-frequency power supply for the counter, a pulse-lengthening multivibrator circuit and a mechanical recorder.

The counter consists of a copper cylinder whose surface is treated with cuprous oxide to increase its photoelectric emission, thus insuring a more uniform discharge current on the incidence of ionizing particles.

Inside the tube is mounted a tungsten wire 20 mils in diameter which points toward a hole in the top of the cylinder. The assembly is mounted inside a glass bulb, with the aperture in the copper cylinder very close to the glass wall. The bulb is filled with a mixture of argon and air. The reel of wire to be tested is placed against the glass bulb, near the opening of the copper cylinder, which enables the ionizing particles to enter the ionizing region near the point of the tungsten wire. This sets off a visible spark and a current pulse each time a particle enters. The pulse is sent on to the counting network. Counting rates of up to 300 per minute are obtainable, and these decrease to four or less on removal of the test wire. The small residual background count is due to stray ionization from radioactive materials or cosmic rays.

G-M COUNTERS

Nuclear Development Laboratory, P. O. Box 7601, Kansas City, Mo. The model B7 bell-type, thin-window counter for measurement of soft beta and alpha radiations is available with uniform mica windows in various thicknesses from 1 mg/cm² and effective diameters of $1\frac{5}{16}$ in., $1\frac{1}{4}$ in., $1\frac{1}{2}$ in. and 2 in. The model BC7 thin-wall, all-metal beta-ray counter can be used to measure beta rays of energy 200 kev or greater and also for X, gamma and cosmic radiation. These counters use a standard col-



lapsible aluminum tube for the cathode shell and have a total wall thickness of 30 mg/cm². The model GC1-7 all-metal, cosmic-ray counter is said to contain a self-quenching gas filling which exhibits unlimited life, and to have a cosmic-ray efficiency greater than 99.5%, no electronegative ion formation which might cause long time lags, and no temperature effects between minus 70° and plus 120° C.

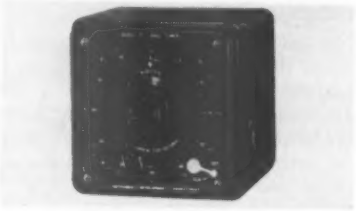
AUTOSCALER

Tracerlab, Inc., 55 Oliver St., Boston, Mass. The SC-1A Autoscaler is said to be an improved design of the first model. The range of the high-voltage

supply has been extended to make it continuously variable from 500–2200 volts and is electronically regulated to 0.01% per percent change in line voltage. A time-delay circuit has been added to control the application of high voltage until all circuit tube filaments have come up to operating temperature. Minor design changes have been made to improve high-voltage stability and to provide a higher safety factor against flashover.

PRECISION TIMER

Instrument Development Laboratories, 229 W. Erie St., Chicago 10, Ill. The model T1 dual timer has been designed



for use with the model 163 and 164 scaling units. The timer will either break an electrical circuit after a preset number of minutes and seconds, or will show elapsed time for an operation. It can be set for any period of time from two seconds to 60 minutes. Although the calibrations on the dial indicate one second, it is claimed that time values as small as ¼ second can be accurately controlled by interpolating between divisions.

THICKNESS GAGE

Goodyear Research Laboratory, Akron, O. The thickness of thin films can be measured in a gage which uses carbon-14 as a source of radiation and an ionization chamber as the detector. The film passes through a slot in the gage. Beta rays from the carbon-14, located below the slot, penetrate the film and produce a current in the detection circuit which is calibrated to read the thickness directly.

ABSTRACTS

CHEMICAL PUBLICATIONS

Some experiments in the preparation of the isotopes U^{239} and Np^{239}, L. Melander (Nobel Inst. Phys., Stockholm), *Acta Chem. Scand.* **1,** 169–177 (1947). The uranyl salicylaldehyde-o-phenylenediimine complex with uranium in pyridine solution was irradiated with neutrons to yield U^{239} and Np^{239} in about 12% yield.

The eight new synthetic elements: G.T. Seaborg (Univ. of California, Berkeley), *Am. Scientist* **36,** 361–376 (1948). A review of the discovery of elements 43, 61, 85, and 87, and of the four trans-uranium elements, with 62 references.

Isotopic exchange and the nature of the chemical bond, M. Haïssinsky, R. Daudel (Inst. du Radium, Paris), *Bull. soc. chim. France* **1947,** 552–559. A review of exchange reactions including a discussion of mechanisms.

Radioactive isotopes as indicators in biology, J. Sacks (Brookhaven Natl. Lab., Upton, N. Y.) *Chem. Rev.* **42,** 411–456 (1948). A critical review of the use of radioactive isotopes in biological research, with 291 references.

Procedure for measurement of P^{31} and P^{32} in plant material, A. J. MacKenzie, L. A. Dean (Dept. of Agriculture, Beltsville, Md.), *Ind. Eng. Chem. Anal. Ed.* **20,** 559 (1948). A routine quantitative procedure for determining the specific activity of plant material containing P^{32} is described in detail. The method comprises digestion of the sample to destroy organic matter and to dehydrate silica, removal of the silica, and precipitation of the phosphorus as magnesium ammonium phosphate hexahydrate. The weight of this precipitate gives the P^{31} content, and a radioactivity measurement yields the P^{32} content.

Radioactive contaminants in tracers, W. E. Cohn (Oak Ridge Natl. Lab., Oak Ridge, Tenn.), *Ind. Eng. Chem. Anal. Ed.* **20,** 498–503 (1948). The origin, detection and identification, and removal of radioactive impurities in tracers are discussed in connection with the principles involved and some specific examples drawn from experience.

Hydrogen exchange of phenols and phenol ethers with deutero-alcohol, P.F. Tryon, W. G. Brown, M. S. Kharasch (Univ. of Chicago, Ill.), *J. Am. Chem. Soc.* **70,** 2003–2005 (1948). The exchange with deutero-alcohol of hydrogen atoms ortho and para to the hydroxyl or alkoxyl group in phenols and phenol ethers was studied. Some exceptions and peculiarities are noted.

The exchange reaction between methane and deuteromethanes on silica-alumina cracking catalysts, G. Parravano, E. F. Hammel, H. S. Taylor (Princeton Univ., Princeton, N. J.), *J. Am. Chem. Soc.* **70,** 2269–2270 (1948). The exchange reaction between methane and deuteromethanes on silica-alumina cracking catalysts was studied, and was found to occur measurably at temperatures of 345° and higher. From these data a mechanism for the cracking process of hydrocarbons is postulated to consist of scission of C—H bonds and then scission of C—C bonds.

Preparation of 1-C^{14}-propene-1 and the mechanism of permanganate oxidation of propene, B. A. Fries, M. Calvin (Univ. of California, Berkeley), *J. Am. Chem. Soc.* **70,** 2235–2240 (1948). Pyrolysis of *n*-propyltrimethylammonium hydroxide gave 1-C^{14}-propene-1 in 90% yield. Other methods of preparation (which are described) led to extensive rearrangement of the double bond. The mechanism of the permanganate oxidation of the labeled propene was investigated and found to depend upon the experimental conditions, particularly the *p*H.

A doubly charged cosmic ray particle. A. B. Sahiar (Tata Inst. Fundamental Research, Bombay, India), *Nature* **161,** 811–812 (1948). Out of a thousand stereoscopic counter-controlled cloud chamber photographs of mesons, one photograph showed that a doubly-charged particle was ejected by a neutral particle accompanying a shower of electrons and mesons.

Anomalous distribution of nuclear disintegration in photographic emulsions exposed to cosmic rays: Double stars, L. Leprince-Ringuet, J. Heidman (L'Ecole Polytechnique, Paris), *Nature* **161,** 844 (1948). An analysis of the distribution of 2,250 nuclear disintegrations in photographic plates exposed to cosmic rays at

a height of 3,600 m shows an anomaly in their distribution. An explanation for this anomaly is proposed.

Anomalous distribution of nuclear disintegration in photographic emulsions exposed to cosmic rays: Double stars, T. T. Li, D. H. Perkins (Imperial College Science and Tech., London), *Nature* **161,** 844–845 (1948). Measurements were made on the surface distribution of 1,230 stars on 54 sq in. of Ilford C2 plates exposed at 3650 m elevation. The observed number of pairs of stars separated by a distance r or less was considerably greater than the expected number, and an explanation is advanced and discussed.

A radioactive local anaesthetic, F. Howarth (Victoria Univ. of Manchester), *Nature* **161,** 857 (1948). Dibromoprocaine may be prepared by the following steps: (*1*) conversion of active bromide (such as KBr) to HBr with hydrogen at 800° C; (*2*) solution of the HBr in water; (*3*) addition of procaine and hydrogen peroxide to the HBr solution. The conversion is almost quantitative and may be completed in about 1.5 hr.

Fractional distillation and its application in the concentration of the heavy isotopes of oxygen and hydrogen, I. Dostrovsky, E. D. Hughes, D. R. Llewellyn (Univ. College of North Wales, Bangor), *Nature* **161,** 858–859 (1948). The construction and operation of a still designed to operate at reduced pressures with either ordinary distilled water or enriched water as feed stock is described. The main section consists of a 31-ft length of 3 in., 16-gage copper pipe, and contains a 30-ft portion filled with 100-mesh phosphor-bronze packing. The boiler has a working volume of 200 ml of water. After 32 days of continuous distillation, a six-fold enrichment of O^{18} was obtained in the boiler water, while the deuterium concentration was about 180 times the normal value. Although at this point it was possible to remove daily 30 ml of enriched H_2O^{18}, this lowered the deuterium enrichment to about 140 times the normal value.

Meson scattering with nuclear excitation, H. J. Bhabha, R. R. Daniel, (Tata Inst. Fundamental Research, Bombay, India), *Nature* **161,** 883–884 (1948). A photographic track that has been obtained appears to be caused by the scattering of a meson by a nucleus in which the nucleus is not disrupted but singly excited. Such a process is possible, but this represents the first experimental evidence that it does actually occur.

Investigation of the radioactivity of rocks by the photographic method, J. H. J. Poole, J. W. Bremner (Trinity College, Dublin), *Nature* **161,** 884–885 (1948). The possibility of estimating the total uranium and thorium content of rocks by placing the flat surface of the specimen cut with a diamond saw against an Ilford nuclear research plate and exposing it was explored. The plates showed that the distribution of the radioactive elements in rocks is sporadic. Corrections must be made for the presence of tracks originating in the photographic plate. Enough data have not yet been obtained to determine uranium or thorium contents.

Mesons and nucleons, O. Klein (Inst. för Mekanik och Matematisk, Stockholm), *Nature* **161,** 897–899 (1948). A speculative discussion is given of the relation between mesons and nucleons. It is assumed that: (*1*) μ-mesons are particles of spin $h/2$ with Fermi statistics and undergo true β decay, (*2*) π-mesons are mestastable compound systems of mesons, (*3*) μ-mesons interact by means of a Yukawa field, and (*4*) nucleons are stable compound systems of mesons. The difficulties as well as agreements arising from these assumptions are discussed. Assumption (*1*) is considered to have an important bearing on the problem of incorporating meson theory into general relativistic field theory.

Excitation curves for the γ-radiation from light elements bombarded with γ-particles from polonium, H. Slätis (Nobel Inst. for Physics, Stockholm), *Nature* **161,** 899–900 (1948). The excitation curves as a function of the energy of the α particles for the γ radiation of lithium, beryllium, boron, carbon, nitrogen, oxygen, fluorine, sodium, magnesium, aluminum, silicon, phosphorus, sulfur, chlorine, potassium, calcium and lead bombarded with α particles from polonium were obtained.

Electron tracks in photographic emulsions, R. H. Herz (Kodak, Ltd., Wealdstone, Middlesex), *Nature* **161,** 928–929 (1948). The maximum energy of electrons which can be recorded with Kodak NT2a emulsion is about 80 kv. NT2a plates were soaked in dilute thorium and uranium nitrate solutions, and electron tracks were recorded at the origin

of α-tracks. Some electron tracks show clumping of grains at the beginning and at the end of tracks; this is probably due to the Auger effect.

Microsynthesis of C^{14}-labeled ethyl iodide, W. J. Arrol, R. Glascock (Univ. of London), *Nature* **161,** 932 (1948). C^{14}-labeled ethyl iodide may be prepared in 70–80% yield on a scale of about 10 micromoles as follows: (*1*) $C^{14}O_2$ is converted to acetylene, (*2*) the latter is converted to ethylene by shaking with freshly prepared chromous chloride solution at room temperature, (*3*) hydrogen iodide is added to the ethylene and the mixture is heated at 144° in a closed tube for one hour.

Riboflavin enhancement of radioactive phosphate exchange by yeasts, W. J. Nickerson, L. J. Mullins (Carlsberg Laboratorium and Inst. for Theoretical Physics, Copenhagen), *Nature* **161,** 939–940 (1948). It has been found possible to increase the radioactive phosphate exchanged by yeast in the presence of glucose by adding trace amounts of riboflavin to the suspending medium. This and other experiments indicate that phosphate is brought through the cell barrier by uniting with some "complexing substance" existing at the cell surface; riboflavin may be such a complexing substance.

Influence of folliculin on bone metabolism, studied by means of radiophosphorus, $_{15}P^{32}$, J. Goraerts, M. J. Dallemagne (Univ. of Liege), *Nature* **161,** 977 (1948). Tracer experiments on pigeons showed that nearly half the phosphorus present in new medullary bone does not come directly from phosphorus that is fed or injected, but from phosphorus already stored in the skeleton. Nearly 13.1% of bone phosphorus was renewed in a pigeon receiving folliculin, while only 1.7% was renewed in the controls.

An apparatus for measuring the energies of fission fragments obtained by slow and fast neutron bombardment, L. Simons, B. Grotenfelt, *Soc. Sci. Fennica Commentationes Phys.-Math.* **13,** 1–9 (1947). An electronic apparatus for measuring the energies of fission fragments is described.

Enrichment of heavy isotopes of lithium and potassium by electrolytic ion migration in fused chlorides, A. Klemm, H. Hintenberger, P. Hoernes, *Z. Naturforsch.* **2A,** 245–249 (1947). Electrolytic ion migration in fused LiCl containing 2 mol % KCl at a current density of 5 amp/cm² for 48 hours led to a change in the ratio Li^7/Li^6 from 12.3 to 44.3, and a change in the ratio K^{41}/K^{39} from 0.0714 to 0.0885.

. . . I. W. RUDERMAN

LIFE SCIENCE PUBLICATIONS

Recording devices in connection with Geiger-Müller counters, T. Kyrre, *Act. Physiol. Scand.* **15,** 11–25 (1948). A summary of various devices for recording the number of impulses received from a Geiger-Müller tube when exposed to radiation from a radioactive preparation. An automatic machine is described which is capable of successively conveying under a G-M tube up to 8 separate preparations once or several times and for periods of time from 2 to 60 minutes.

Isotopes in medicine, G. Medes (Lankenau Hospital Res. Inst. and Inst. for Cancer Res., Philadelphia, Pa.), *Am. J. Clin. Path.* **18,** 354–361 (1948). A review of the use of isotopes in physiology and medicine. Radioactive and stable isotopes are considered. Forty-six references are cited.

The effect of beta rays on cells cultivated *in vitro,* I. Lasnitzki (Strangeways Res. Lab., Cambridge, England), *Brit. J. Radiology* **21,** 265–269 (1948). Avian fibroplast tissue cultures were exposed to doses of beta rays. Mitosis alteration and the production of abnormal mitotic and degenerate cells were examined for a period of one day after exposure to radiation. Shortly after exposure, a steep fall in mitosis and the appearance of abnormal mitotic figures were observed. This was followed by a return of mitotic activity accompanied by a more abnormal mitosis and cell degeneration. At the end of the observation period mitotic recovery was half the normal value.

Tissue cultures treated with the same dose of X-rays showed the same qualitative effects and the total damage was of the same order. The time relation, however, differed markedly in the two experiments. Tissue cultures irradiated with beta rays show the greatest effect shortly after exposure and recovered sooner. After exposure to X-rays, the radiation effect was delayed and recovery was slow.

Plain radiography of the skull in the diagnosis of intracranial tumors, D. Stenhouse (Killearn Hosp., Stirlingshire, Scotland), *Brit. J. Radiology* **21,** 287–300 (1948). An analysis is made of radiographic findings in 200 verified cases of intracranial tumors. Of all the abnormal sellar appearances, only two were considered to be of localizing values: uniform expansion with little or no thinning of dorsum, and suprasellar calcification. The commonest changes were thinning or erosion of dorsum. The degree of sellar destruction and the degree of hydrocephalus in metasellar lesions showing changes are roughly proportionate to one other, but the hydrocephalus in itself is not the cause of the bone destruction.

In 22% of all cases, signs of increased intracranial tension comprising starting of sutures, increased convolutional markings, widening of the occipital emissary foramina and thinning or erosion of dorsum sellae were found. In 30.5%, localizing signs were found.

Some aspects of radiology in war-time Germany, G. Donovan (Publ. Health Dept., Gorseinon, Glam., Wales), *Brit. J. Radiology* **21,** 305–308 (1948). Trends in German radiology are toward the use of automatic methods, compensating devices, safety measures and foolproof apparatus. It is doubtful whether the war significantly contributed to the development of German radiology.

A simple apparatus for making rapid serial radiographs of small objects, G. Foxon (Guy's Hospital Med. School), *Brit. J. Radiology* **21,** 301–304 (1948). An apparatus is described for taking serial radiographs of small fields up to the rate of three per second. A continuous beam of X-rays is interrupted by a shutter placed above the object being radiographed; while the shutter is closed a film placed below the object moves forward and then becomes stationary while the next exposure is made.

Radioactivity distribution in the tissues of mice bearing melanosarcoma after administration of D,L-tyrosine labeled with radioactive carbon, J. Reid, H. Jones (Radiation Lab., Dept. of Chem., Univ. of Calif., Berkeley), *J. Biol. Chem.* **174,** 427–437 (1948). D,L-tyrosine labeled with C^{14} in the beta position was administered intravenously to mice bearing melanosarcoma. The distribution of radioactivity was studied. At the end of 72 hours 30% of the administered dose appeared in the breath, about 40% appeared in the urine and feces and about 30% remained in the body. Every tissue contained some radioactivity. The highest specific activities were found in the adrenals, thyroids, intestines, kidneys, liver, plasma, spleen and tumor. Erythrocytes and bone had the lowest specific activities. The total activity in the body was diminished by half in the 60-hour period between the 12th and 72d hour after administration of radiotyrosine.

Plasma inorganic iodide as a homeostatic regulator of thyroid function, J. Wolff, I. Chaikoff (Div. of Physiology, Univ. of California Med. School, Berkeley), *J. Biol. Chem.* **174,** 555–564 (1948). Rats were injected with varying amounts of radioactive iodine in the form of KI. The radioactivity served as a measure of injected I^{127} that entered the thyroid. As long as the plasma iodide was above 20 to 35 gamma percent, the organic binding of the injected iodide was blocked. A homeostatic mechanism is presented which postulates that excessively high levels of plasma iodine inhibit formation of the hormone in the thyroid gland. This helps to explain the beneficial effect produced in patients suffering from Graves' disease when large amounts of iodine are administered. The thyroid gland is still able to concentrate iodine when the organic binding of iodine is blocked. These results establish that the normal thyroid gland possesses a mechanism for concentrating iodine which is not dependent upon its conversion to thyroxine and diiodotyrosine.

Observation on the utilization of glycine in the biosynthesis of hemoglobin, M. Grinstein, M. Kamen, C. Moore (Dept. of Internal Med. and Mallinckrodt Inst. of Radiol., Washington Univ. School of Med., St. Louis), *J. Biol. Chem.* **174,** 767–768 (1948). Glycine labeled with C^{14} in the carboxyl group was administered to a dog via stomach tube. Blood samples were obtained at various intervals and crystalline protoporphyrin methyl ester and globin were isolated and analyzed for radioactivity. No radioactivity was observed in the protoporphyrin. Globin radioactivity was constant throughout the observation period. The results show that the carboxyl carbon of glycine is not utilized in protoporphyrin synthesis but is incorporated into the globin molecule.

The effect of dinitrophenol on the incorporation of alanine labeled

with radioactive carbon into the proteins of slices of normal and malignant rat liver, I. Frantz, Jr., P. Zamacnik, J. Reese, M. Stephenson (Med. Lab. of the Collis P. Huntington Memorial Hosp. of Harvard Univ., at Massachusetts General Hosp., Boston), *J. Biol. Chem.* **174,** 773–774 1948. It was found that addition of dinitrophenol to rat liver slices incubated in Krebs-Ringer-phosphate medium with tagged D,L-alanine inhibits the incorporation of alanine into protein in both normal and tumor tissue, while permitting oxygen consumption to continue. This suggests that the synthesis of alanine peptide bonds depends on a source of energy-rich phosphate donors. Results showed a higher rate of incorporation of alanine into hepatoma protein than into normal liver protein when dinitrophenol was not used.

. . . BERNARD KANNER

PHYSICAL PUBLICATIONS

On the space energy distribution of neutrons in a moderator of infinite size, G. Holtz, *Arkiv Mat., Astron. Fysik* (Stockholm) **35,** No. 2, 1–12 (1948). Assuming isotropic emission from a small volume, isotropy of scattering in the center of gravity system, and a simple variation of the mean free path with the neutron energy, a formula for the distribution function of neutrons in space and energy is derived, and from it the second and fourth moments of the distribution function are calculated.

On the β- and γ-radiation from U^{239} and Np^{239}, H. Slätis, *Arkiv Mat., Astron. Fysik* (Stockholm) **35,** No. 3, 1–23 (1948). A uranium target was bombarded with deuterons and the radiations subjected to measurement by absorbers, showing that U^{239} has two gamma components of energies 920 and 83 kev. Np^{239} gives off 500, 200, and 51 kev gammas with a 2.3-day period. A magnetic lens spectrometer measured a continuous spectrum for U^{239} betas which is complex, having components of maximum energies 1.12 and 2.06 Mev. The beta spectrum of Np^{239} shows conversion lines from six gamma rays and four groups of electrons. Conversion coefficients are calculated and disintegration schemes suggested.

A high-frequency cyclotron generator with demountable tubes, H. Atterling, G. Lindström, *Arkiv Mat., Astron. Fysik* (Stockholm) **35,** No. 4, 1–9 (1948). An oscillator is described which gives a high efficiency in the r.f. circuit. Diode voltmeters give grid and plate voltages useful for tuning, and tube filaments are protected by a vacuum interlock.

An equipment for automatic resonance control, G. Lindström, *Arkiv Mat., Astron. Fysik* (Stockholm) **35,** No. 5, 1–8 (1948). An automatic mechanism is described that changes tuning of the r.f. circuit automatically to compensate for drifts in the magnetic field of a cyclotron to keep the magnetic resonance condition operative for the accelerated ions.

On the role of the exchange forces in the nuclear three- and four-body problems, N. Svartholm, *Arkiv Mat., Astron. Fysik* (Stockholm) **35,** No. 7, 1–7 (1948). Using the method of iterated functions in the momentum space, the energy levels of such problems are calculated with nearly correct symmetry properties for the wave functions.

Industrial application of Geiger-Müller counters to the analysis of phosphorus in steels, F. R. Bryan, G. A. Nahstott (Ford Motor Co., Dearborn, Mich.), *J. Optical Soc. Am.* **38,** 510–517 (1948). A Littrow temperature-controlled quartz spectrograph directs spectral lines of various wavelength on two G-M counters which, by counting photons, determine the phosphorus content of production samples in open-hearth and foundry furnace production.

Compact electrostatic generator for the production of positive ions, W. E. Danforth, E. L. Hudspeth, C. E. Mandeville, C. P. Swann (Bartol Research Foundation, Franklin Inst., Swarthmore, Pa.), *Phys. Rev.* **73,** 1413–1418 (1948). A pressure-insulated generator of compact design, utilizing a low voltage arc to provide ions, is described. The accelerating tube withstands the developed voltage of 1.5-million volts across its length of five feet. Positive ion beam voltages of 150–1750 kev have been used, with magnetic analysis providing 0.1 to 2 microamperes of resolved beam.

Cosmic-ray investigations on Mt. McKinley, T. D. Carr, M. Schein, I. Barbour (Univ. of Chicago, Ill)., *Phys. Rev.* **73,** 1419–1423 (1948). An experi-

ment on the absorption of penetrating particles in lead, performed at 18,000 feet indicates large numbers of mesotrons of about 200 Mev. Comparison with measurements at 14,250 feet indicates that production of slow mesotrons (under 400 Mev) takes place between the two altitudes. Using paraffin and lead blocks with coincidence circuits, an upper limit of 3% is set to mesotrons capable of penetrating 10 cm of Pb produced by nonionizing radiation in up to 10 cm of Pb and paraffin.

The beta- and gamma-rays of Ga^{72}, A. C. G. Mitchell, D. J. Zaffarano, B. D. Kern (Indiana Univ., Bloomington), *Phys. Rev.* **73,** 1424–1429 (1948). The spectrum of Ga^{72} (14.1-hr half-life) was investigated with a magnetic lens spectrometer. Photoelectrons from a thin lead radiator provided the gamma-ray spectrum with strong lines at 0.631, 0.835, 1.05, 2.18, and 2.50 Mev. Some weaker lines were also found. A Fermi plot of the beta spectrum indicated exceeding complexity, with seven groups of electrons suggested, having endpoints at 3.17, 2.57, 1.74, 1.45, 1.00, 0.74, and 0.56 Mev. A tentative disintegration scheme is proposed, containing eight levels in Ge^{72}.

A study of the radiations from iridium (194), iridium (192), lanthanum (140), antimony (124), and zirconium (95), C. E. Mandeville, M. V. Scherb (Bartol Research Foundation, Franklin Inst., Swarthmore, Pa.), *Phys. Rev.* **73,** 1434–1441 (1948). Absorption and coincidence measurements were made on radioactive isotopes of these elements as produced by neutron bombardment in the Clinton pile. The beta- and gamma-ray energies are given along with the coincidence rates, and disintegration schemes given in some cases.

Artificially radioactive Se^{73} and Se^{75}, W. S. Cowart, M. L. Pool, D. A. McCown, L. L. Woodward (Ohio State Univ., Columbus), *Phys. Rev.* **73,** 1454–1457 (1948). Alpha-particle bombardment of Ge produces a 7.1 hour activity ascribed to Se^{73}. Absorption measurements indicate a positron endpoint of 1.29 Mev, and X-radiation of 1.17 A.U. This activity is stronger when Ge enriched in Ge^{70} is used. The 127-day Se^{75} activity gives gamma rays at 0.22 and 0.43 Mev.

Range of protons from $N^{14}(n,p)C^{14}$, I. C. Cornog, W. Franzen, W. E. Stephens (Univ. of Penn., Philadelphia), *Phys. Rev.* **74,** 1–4 (1948). An 800 mc Ra-Be neutron source was surrounded by paraffin and placed near a cloud chamber containing nitrogen, *n*-propyl alcohol, and water to a total pressure of one atmosphere. The stopping power of the gas was determined by observing the range of Po alpha particles. Photographs of the protons from the reaction give a corrected range of 0.996 cm. The energy of these protons as calculated using the C^{14} disintegration energy gives 561 kev – 1.00 cm as a point on the range-energy curve for protons.

The emission of protons from light nuclei on bombardment with α-particles of short range, F. C. Champion, R. R. Roy (Univ. of London, Eng.), *Phys. Rev.* **74,** 5–8 (1948). Cloud chamber pictures were taken of (α,p) reactions on fluorine, neon, and nitrogen. Protons of a single group were found for neon (indicating a ground state), while protons from fluorine varied widely in range. The protons were emitted anisotropically in fluorine and nitrogen, and it is indicated that the incident particles had angular momentum about the center. Relative reaction probabilities were found and an explanation is suggested.

Theory of the lithium two-alpha-reactions—I. Angular distribution of $Li^7(p,\alpha)\alpha$, D. R. Inglis (Johns Hopkins Univ., Baltimore, Md.), *Phys. Rev.* **74,** 21–33(1948). Because the resulting two alpha particles of this reaction have even parity, only the *p* and *f* protons (disregarding higher angular momenta) need be considered, and the angular distribution is calculable on the basis of dispersion theory. Compound nuclear levels of angular momentum 0 and 2 were considered. Coefficients (as functions of energy) were derived for the $\cos^2$ and $\cos^4$ terms in the angular factor, and fitted well with experimental data when both *p* and *f* waves were included. More accurate data may require the assumption of an additional compound level.

Energy quanta emitted in disintegration of ground state of Ba^{133}, F. C. Yu, J. D. Kurbatov (Ohio State Univ., Columbus), *Phys. Rev.* **74,** 34–37 (1948). Barium bombarded with slow neutrons shows four periods, and two gamma-ray energies were established. These result

from the two excited states in Cs^{133*} after K-capture takes place in radioactive barium. The ratio of the number of 320-kev quanta to 85-kev quanta is about 1.31.

Experiments on the separation of low energy mesotrons from electrons, P. C. Bhattacharya (Calcutta Univ., India), *Phys. Rev.* **74,** 38–43 (1948). A lead block 1.25 cm thick was used to produce showers below the topmost counter of a telescope, the showers then being suppressed by anticoincidence counters below. This eliminated electrons that produce showers; those which do not (low-energy ones) were absorbed in a second lead block below the anticounters but above the lowest counter of the telescope. Mesotrons of energy as low as 1.6×10^8 ev could pass through the telescope, and be detected in a cloud chamber following it. Experiments were carried out on the number of showers of various numbers of particles produced by electrons in various thicknesses of lead.

Radioactive europium, gadolinium, and terbium, N. L. Krisberg, M. L. Pool (Ohio State Univ., Columbus), C. T. Hibdon (Univ. of Colorado, Boulder), *Phys. Rev.* **74,** 44–47 (1948). Neutron bombardment of gadolinium results in 18-hr and 5.5-day activities. The 18-hr activity decays by negatron emission of 0.85 Mev energy and gamma-ray emission of 0.3 Mev. The 5.5-day activity decays by negatron emission of 0.5 Mev and gamma-ray emission of 1.28 Mev. The mass assigned to both activities is 161. Deuteron bombardment of europium produces activities of 75 days and 20 years, the latter decaying with a 1.4-Mev beta ray and a gamma ray.

Radiation from electrons accelerated in a synchrotron, F. R. Elder, R. V. Langmuir, H. C. Pollock (General Electric Co., Schenectady, N. Y.), *Phys. Rev.* **74,** 52–56 (1948). Spectral distribution of radiation (almost entirely caused by the continuous centripetal acceleration) during the acceleration of electrons to several different levels of peak energy has been observed. The distribution was that of an electron of energy E which varied sinusoidally. A spectroradiometer which automatically records the spectra shows excellent agreement with theory, the deviations observed probably being caused by fluctuations of electron-beam current during a run.

Radioactive isotopes of Ru and Tc, D. T. Eggen, M. L. Pool (Ohio State Univ., Columbus), *Phys. Rev.* **74,** 57–62 (1948). Bombardment of Mo with alpha particles, Ru with neutrons, Mo with protons, and Mo^{94} with alphas allowed assignment of 1.65-hr, 20-hr, and 2.8-day activities to Ru^{95}, Tc^{95}, and Ru^{97}, respectively. Ru gave off positrons of 1.1 Mev, X-rays of 0.7 A.U., and gammas of 0.95 Mev. Tc gave off the 0.7 A.U. X-rays and a 0.78 Mev gamma ray. The technetium region of the isotope chart is given.

Mass temperature coefficients of cosmic-ray components, F. M. Millican, D. H. Loughridge (Univ. of Washington, Seattle, Wash.), *Phys. Rev.* **74,** 66–70 (1948). An average temperature of the atmosphere is calculated by weighting the temperatures at different levels according to their pressures. The theory of the temperature coefficient of cosmic-ray intensity is given and compared with the experiments.

Molecular spectroscopic evidence of the existence of strontium isotopes Sr^{84}, Sr^{87}, and Sr^{86}, L. H. Ahrens (Massachusetts Inst. of Technology, Cambridge), *Phys. Rev.* **74,** 74–77 (1948). Study of band heads in SrF arc emission spectra shows three components, whose displacements indicate the presence of three strontium isotopes of masses 88, 87, and 86. The abundance ratio of 88/86 determined from the spectra agrees to 2.0% with that from mass spectrometers; the ratios involving mass 87 are only approximate because of its masking by mass 88, but this would not be the case in a higher order. Geological age requirements are discussed, the conclusion being drawn that spectroscopic determinations require too much strontium salt.

Internal conversion electrons from Sm^{147}-47 hour, R. D. Hill (Univ. of Illinois, Urbana), *Phys. Rev.* **74,** 78–79 (1948). A beta-ray spectrograph of the semicircular focusing type was used, with a photographic plate as a detector, to find the spectrum of internal conversion lines from Sm^{153}. Nine lines are assigned to conversion in various shells of two gamma rays of 103 and 69.5-kev energy. Some lines are Auger lines from the conversion of Eu X-rays following K-capture.

Waves and electrons traveling together —a comparison between traveling wave tubes and linear accelerators, L. Brillouin (Harvard Univ., Cambridge, Mass.), *Phys. Rev.* **74,** 90–92 (1948).

The two devices transfer energy from the waves to the electrons or vice versa, depending on the space charge, power level of the waves, and change in the wave velocity. A general discussion of both electrons and waves traveling together is included, and a special case treated.

On the neutron-proton force, J. M. Blatt (Massachusetts Inst. of Technology, Cambridge), *Phys. Rev.* **74,** 92–96 (1948). Low-energy scattering data do not give information on the shape of the well but can be analyzed to find the ranges in the triplet and singlet states without knowing the well shape. Singlet and triplet plots show that scattering experiments alone are not enough to determine even the effective ranges of the *n-p* forces in the two spin states, but give a functional relation between them. More information can be obtained from a direct measurement of the coherent *n-p* scattering amplitude at low energies. Together with the epithermal cross section, it allows the conclusion that if the nuclear potential is not "long-tailed," the effective triplet range is from 1.30–1.55 $\times 10^{-13}$ cm, and the effective singlet range is less than 2.0 $\times 10^{-13}$ cm, in contradiction to the hypothesis of the charge-independence of nuclear forces.

A further study of the γ-radiation from polonium, B. Zajac, E. Broda, N. Feather (Univ. of Edinburgh, Scotland), *Proc. Phys. Soc.* **60,** 501–508 (1948). Polonium emits soft γ-rays, the most intense of which (taking place in 0.001% of the disintegrations) has energy of 84 ± 4 kev, as determined by absorption in lead, gold, and tungsten. That they are not due to disintegration effects or inelastic scattering of alpha particles is shown by their persistence when 50 mg/cm² of polythene is placed over the source. The delay of gamma emission is less than 0.1 sec. These data do not explain those on alpha-particle fine structure.

The semi-diurnal variation in cosmic ray intensity, P. Nicolson, V. Sarabhai (Cavendish Lab., Cambridge, England), *Proc. Roy. Soc.* **60,** 509–523 (1948). The 12-hour period of variation in the intensity of the energetic meson component at sea level is discussed. It shows a strong correlation with atmospheric pressure variations, indicating that atmospheric oscillations may be responsible. This theory is rejected because it would quantitatively indicate major production of mesons at 60 km height, which corresponds to an inordinately high cross section for meson production.

The neutrons emitted in the disintegration of nitrogen by deuterons, W. M. Gibson, D. L. Livesey (Cavendish Lab., Cambridge, Eng.), *Proc. Roy. Soc.* **60,** 523–532 (1948). Neutrons from the reaction $N^{14}(d,n)O^{15}$ were detected by proton recoils in photographic plates. Aluminum nitride targets were used, and the deuteron energy was 930 kev. A Q value of 5.15 ± 0.10 Mev was deduced from the neutron energies, no lower energy group of neutrons being observed of an intensity as high as 15% of that of the above group. The angular distribution of the neutron yield was also compared with that of D-D neutrons.

Efficiency of counting systems, M. Blackman, J. L. Michiels (Imperial College, London, England), *Proc. Roy. Soc.* **60,** 549–561 (1948). Formulae are derived for the efficiency of a counting system including a counter, an amplifier, a scaling circuit, and a recorder (including a recorder of constant resolving time). Previously derived formulae are investigated and some discarded (for instance, a choice is made among three proposed formulae giving the counting loss due to the dead-time of the counter). Examples are given of the use of these formulae in various counting experiments.

Geometric factors underlying coincidence counting with Geiger counters, H. E. Newell, Jr. (Naval Research Lab., Washington, D. C.), *Rev. Sci. Instr.* **19,** 384–389 (1948). The coincidence rate of a two-counter telescope (perfectly efficient) in an isotropic field of radiation is a function of its intensity and the dimensions and separation of the counters. Graphs are given showing upper bound of the coincidence rate as a function of the ratio of the diameter to the separation of the counters, and of the ratio of the length to the separation. Curves showing the maximum error in the previous graphs are also given.

An inside Geiger-Müller counter for soft beta-emitters, L. W. Labaw (National Inst. of Health, Bethesda, Md.), *Rev. Sci. Instr.* **19,** 390–395 (1948). A working inside counter giving high counting rate and reproducible results is described, together with the effects of self-absorption in the emitter. A method of radioactive assay is also given.

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